THÈSE

En vue de l'obtention du

DOCTORAT DE L'UNIVERSITÉ DE TOULOUSE

Délivré par :

Institut National Polytechnique de Toulouse (INP Toulouse)

Discipline ou spécialité :

Sciences des Agroressources

Présentée et soutenue par :

M. PABLO LOPEZ HURTADO

le jeudi 8 septembre 2016

Titre :

DEVELOPMENT OF NOVEL BUILDING INSULATION MATERIALS, INCORPORATING CELLULOSE AND BIOBASED ADDITIVES

Ecole doctorale :

Sciences de la Matière (SDM)

Unité de recherche :

Laboratoire de Chimie Agro-Industrielle (L.C.A.)

Directeur(s) de Thèse :

M. LUC RIGAL

MME VIRGINIE VANDENBOSSCHE

Rapporteurs :

M. CHRISTOPHE BALEY, UNIVERSITE DE BRETAGNE SUD

M. SOFIANE AMZIANE, UNIVERSITE BLAISE PASCAL

Membre(s) du jury :

M. BERNARD PERRIN, INSA TOULOUSE, Président

Mme VIRGINIE VANDENBOSSCHE, INP TOULOUSE, Membre

ABSTRACT

Cellulose insulation is manufactured from recycled paper fibres, treated with mineral additives acting as flame retardants and antifungals. Its consistency is similar to cotton wool. The fibres are sold in bulk to be blown into the walls and attics. Its thermal conductivity is around 0.04 W/m.K, which is comparable to glass wool, but it is made with recycled materials and has much lower embodied energy levels. It can be either blown dry or sprayed with water.

The wet spray method for cellulose insulation has several benefits compared to the dry process. Since the cellulose fibres become rigid after drying, it prevents the compaction of the material thus avoiding thermal bridges in the building envelope. However, the time to reach the dry state may be very long and variable depending on the dosage used and the environmental conditions of application. There are many bio-based additives that can contribute to the reduction of this period and improve the cohesion of the material. This research project aims to find the optimal additive for this application while retaining the favourable properties of the insulating material.

Two cellulose types have been characterized with regards to the properties of the fibres to determine their performance with water. Both samples showed differences in chemical composition, grain size, and morphology. The values of water retention, water adsorption isotherms and the proportions of free and bound water have been factors which have shown an influence on the drying of the insulation. Density, compressive strength, and thermal conductivity increased with moisture dosage. A minimum of 14 kPa for the compression module was defined as the mechanical property threshold to avoid settling. These properties were compared with those of the cellulose insulation compacted to dryness and the results showed the strong influence of the stiffening and pore closing process upon drying, on these properties.

Potential bio-based additives were classified and characterized with regards between concentration, viscosity, and adhesive strength. A relationship between these parameters was established. Most showed Newtonian behaviour at low concentrations, with some non-Newtonian concentrations having a pumpable viscosity. Unfortunately several additives which showed good adhesive properties were too viscous and vice versa. A range of surfactants were also considered. Sprayable formulations were characterized with respect to their drying time, compressive strength and thermal conductivity. Additives which have shown positive contributions drying are the lignosulfonate and the cationic surfactant CTAB.

The influence of these additives on drying, with varying weather, liquid dosage and thickness of insulation was defined with a numerical model through the WUFI software. Optimal conditions in which the lignosulfonate additive is more effective have been defined. A first assessment of the performance of new formulation in terms of fire and mould was made and indications for the continuation of the study of the material were formalized.

RESUME

La ouate de cellulose utilisée pour l'isolation est fabriquée à partir de fibres de papier broyé, traitées avec des additifs minéraux agissant comme agents ignifuges et antifongiques. La conductivité thermique du matériau final est aux alentours de 0,04 W/m.K, ce qui est comparable à la laine de verre, avec l'intérêt d'être fabriqué à partir de matériaux recyclés, représentant un taux d'énergie grise beaucoup plus faible.
Le mode de mise en œuvre par voie humide de la ouate de cellulose a plusieurs avantages par rapport à la voie sèche. Le fait que les fibres de cellulose se rigidifient avec l'eau, empêche le tassement du matériau, qui peut engendrer des ponts thermiques dans l'enveloppe du bâtiment. Par contre, la durée de séchage peut être très longue et variable selon le dosage utilisé et les conditions ambiantes d'application. Ce projet de recherche vise à trouver l'additif idéal permettant d'accélérer le séchage tout en conservant une bonne cohésion du matériau et le maintien de ses propriétés isolantes.
Deux types de ouate de cellulose utilisés en isolation ont été caractérisés. Ils ont montré des différences de composition chimique, granulométrie et morphologie. L'influence de leurs caractéristiques physiques telles que la rétention d'eau, les isothermes d'adsorption d'eau et les proportions d'eau libre et liée sur le séchage du matériau final a été mise en évidence. Du point de vue de la mise en œuvre, il a été démontré que le dosage en eau avait un impact important sur les propriétés finales du matériau. La densité, la résistance en compression et la conductivité thermique augmentent avec le dosage en eau. Un minimum de 14 kPa pour le module de compression a été défini comme le seuil de résistance permettant d'éviter le tassement. Ces propriétés ont été comparées avec celles de la ouate de cellulose compactée à sec et les résultats ont montré la forte influence de la rigidification et de la fermeture des pores du matériau.
Deux voies ont été envisagées pour résoudre le problème du temps de séchage : l'ajout d'additifs aux propriétés adhésives permettant de réduire la quantité d'eau introduite en renforçant la cohésion de l'isolant, et l'ajout d'additifs permettant de modifier la tension de surface pour faciliter le départ de l'eau. Les additifs biosourcés potentiels ont été caractérisés à différentes concentrations et classés selon leur viscosité et leur pouvoir collant. Malheureusement plusieurs additifs ont dû être rejeté car ils présentaient un couple « propriété adhésive/pompabilité » non adapté. Une gamme de tensioactifs a également été testée par rapport à leurs tensions de surface. Les formulations pompables ont étés caractérisées par rapport à leurs temps de séchage, résistance en compression et conductivité thermique. Les additifs qui ont montré des contributions positives sur le séchage sont les lignosulfonates et le tensioactif cationique CTAB.
L'influence de ces deux additifs a ensuite été étudiée avec un modèle numérique à travers le logiciel WUFI en prenant en compte l'impact sur le séchage, la météo, le dosage liquide et l'épaisseur de l'isolant. Les conditions optimales ont été définies. Le lignosulfonate s'est avéré être l'additif le plus efficace. Une première évaluation de la performance des nouveaux isolants en termes de résistance au feu et à la moisissure a été réalisée et des indications pour la suite de l'étude ont été proposées.

REMERCIEMENTS

Dans mon projet de thèse, l'intervention et dévouement de plusieurs personnes m'ont permis non seulement de réaliser les objectifs de la thèse mais aussi de me permettre d'évoluer comme chercheur.

Je remercie tout d'abord mes directeurs et encadrants de thèse. M Luc RIGAL. Je vous remercie d'avoir accepté d'être mon directeur de thèse et pour votre accueil au sein Laboratoire de Chimie agro-industriel. Je remercie M. Antoine Rouilly et Mme Virginie VANDENNBOSSCHE, pour leur direction, et leur confiance tout au long de ces années de recherche. Vous m'avez également permis de découvrir dans de bonnes conditions le métier de chercheur au travers de vos ambitions et de vos investissements pour développer le projet de recherche.

Je voudrais également remercier les membres du jury : M Christophe BALEY, M Sofiane AMZIANE, et M Bernard PERRIN, pour avoir accepté de faire partie de mon jury de thèse. Je tiens également à leur remercier pour toutes les remarques et discussions constructives qu'on a pu avoir.

Je remercie également mon directeur industriel, le président de Greenbuilding, M. Frank MIRAUX, pour m'avoir accepté au sein de Greenbuilding et pour m'avoir confié cette mission R&D. Je remercie aussi Gilles AMBLARD pour ses conseils sur la ouate de cellulose projetée.

La contribution de Christine RAYNAUD, directrice du CRITT a été essentielle pour établir le lien entre le laboratoire et l'entreprise.

Au sein du Laboratoire de Chimie Agro-industrielle de l'INP – ENSIACET de Toulouse, plusieurs personnes m'ont aidé pour accomplir la thèse. Tout d'abord je remercie les directeurs Gérard VILAREM et Carlos VACA GARCIA pour m'avoir accueilli au sein du laboratoire. Je remercie l'aide et la disponibilité de toute l'équipe du laboratoire, notamment l'équipe logistique : Romain ARRMENGAU, Didier DANGLA, et Gilles HUMBERT. Pour la formation des manipulations expérimentales, Anne LUNG, Guadalupe VACA GARCIA, et Jean-François FABRE ont été indispensables. Je

remercie aussi tout l'équipe de soutien à la recherche, notamment Isabelle BEAUVIES, et Karine TREMAUVILLE.

J'adresse toute ma gratitude à mes camarades thésards qui m'ont tant soutenu, encouragé et fait rire aussi. Il y a les « anciens », avec qui j'ai vécu ces trois années ou la grande partie : Cécile, Alice, Benjamin, Jeremy, Bastien, et Sabina, les « latinos » qui m'ont permis de me détendre dans ma langue maternelle : Marian, Nydia, Jesus, Nati, Isaura, et Amaranta, et les « petits nouveaux » auxquelles je souhaite rencontrer plus dans la suite : Etienne, Rémi, Nico, et Francisco. Vous m'avez tous aidé survivre la thèse.

Je ne souhaite pas oublier de remercier mon stagiaire Thibaut VINE, qui m'a beaucoup aidé sur la caractérisation des additifs biosourcés.

Je remercie mes copains de Mexique : Pollo, Vergara, Crown, et Ana, qui m'ont toujours motivé avec des messages d'encouragement, même de très loin.

Je remercie ma famille qui a su m'apporter encouragements depuis le Mexique, même en moments de frustration. Mama, Papa, Hermanita, sin ustedes, todo esto no lo hubiera podido hacer.

Enfin je remercie à Amy, ma copine, qui m'a supporté pendant les moments difficiles de la rédaction, Les longues journées et soirées de thèse ont été beaucoup plus faciles avec toi.

TABLE OF CONTENTS

Introduction **1**

Chapter 1: State of the art *4*

1.1 Introduction, context and objectives 4

1.1.1 Sustainability in the building sector 4

1.1.2 Cellulose fibre insulation (CFI): a sustainable solution 5

1.2 The properties of cellulose fibre insulation (CFI) 7

1.2.1 Composition 7

1.2.2 Production 8

1.2.3 Installation of CFI 9

1.2.4 Density and settling 10

1.2.5 Thermal properties 12

1.2.6 Air infiltration 15

1.2.7 Fire properties: 15

1.2.8 Fungal development 16

1.2.9 Life Cycle Analysis 17

1.2.10 Moisture properties 19

1.2.11 Drying of wet spray CFI 21

1.2.12 Present opportunities in cellulose insulation research 22

1.3 Biobased additives and interactions with cellulose 23

1.3.1 Adhesives: an introduction 23

1.3.2 Mechanisms of adhesion 24

1.3.3 Adhesion in cellulose fibres 27

1.3.4 Types of adhesives in natural fibre composites 29

1.3.5 Biobased adhesives 30

1.3.5.1 Cellulose esters 30

1.3.5.2 Chitin and chitosan 31

1.3.5.3 Pectins: 32

1.3.5.4 Natural Gums 33

1.3.5.5 Starch 34

1.3.5.6 Lignin 35

1.3.5.7 Tannin: 36

1.3.5.8 Protein glues 37
1.3.6 Surfactants 40
1.3.6.1 Types of surfactants 40
1.3.6.2 Surfactants as dewatering agents in cellulose 41
1.4 Conclusions 42
Chapter 2: The properties of cellulose insulation and the their relation to water dosage 46
2.1 Introduction 46
2.2 Properties of cellulose insulation fibres 47
2.2.1 Origin and preparation of raw materials 47
2.2.2 Microscopic observation 48
2.2.3 Schopper Reigler freeness testing 49
2.2.4 Tapped density and loose fill settling 50
2.2.5 Particle size distribution 52
2.2.6 Chemical composition analysis: 53
2.2.7 Interactions between water and cellulose insulation 54
2.2.7.1 Water retention value 54
2.2.7.2 Bound water determination with differential scanning calorimetry (DSC) 56
2.2.7.3 Dynamic vapour sorption (DVS) 58
2.2.8 Drying of cellulose samples 59
2.2.8.1 Influence of treatment and source material 60
2.2.8.2 Influence of installed water dosage on drying 62
2.2.9 Changes in density 63
2.2.10 Mechanical testing 64
2.2.11 Thermal conductivity testing 69
2.3 Conclusions 72
Chapter 3: Biobased additives and their incorporation with wet spray cellulose insulation 74
3.1 Introduction 74
3.2 Biobased Adhesives 76
3.2.1 Formulation and preparation 76
3.2.2 Viscosity of biobased adhesive solutions 78
3.2.3 Tensile strength 83
3.2.4 Compatibility with spraying equipment 87
3.3 Surfactants 89
3.3.1 Formulation 89

3.3.2 Surface Tension 90
3.3.3 Compatibility with spraying material 91
3.4 Influence of additives on the properties of CFI 91
3.4.2 Drying via binder evaporation and solids content increase 91
3.4.3 Drying via surface tension reduction 94
3.4.4 Mechanical resistance and density 95
3.4.5 Drying of reduced moisture content samples 99
3.4.6 Thermal conductivity 100
3.5 Conclusions 101
Chapter 4: Final properties of insulating material and hygrothermal modelling 104
4.1 Introduction 104
4.2 Influence of additive dosage on the properties of cellulose insulation 105
4.2.1 Drying 105
4.2.2 Compression resistance 107
4.2.3 Thermal conductivity 108
4.3 Calculated Drying performance of treated and untreated CFI 109
4.3.1 Empirical model 110
4.3.1.1 Influence of sprayed moisture. 111
4.3.1.2 Influence of insulation thickness 113
4.3.1.3 Influence of additives. 115
4.3.2 Numerical model: WUFI software 116
4.3.2.1 Theory 116
4.3.2.2 Input parameters 117
4.3.2.3 Specific heat capacity 118
4.3.2.4 Liquid transport coefficients. 118
4.3.2.5 Validation of parameters 121
4.3.3 Drying performance of CFI under different installation conditions 123
4.4 Final product considerations 129
4.4.1 Fire resistance measurements 129
4.4.1.1 Smouldering tests 129
4.4.1.2 Open flame Test 130
4.4.2 Fungal resistance observations 132
4.4.3 Final product properties and further material studies 134
4.5 Conclusions 138

General conclusions and perspectives 140

Experimental Methods 145

List of Figures 168

List of Tables 172

Bibliography 174

Resumé en français 185

INTRODUCTION

Insulation materials are an essential part of a building envelope. They assure that the inside temperature of a room is maintained at a certain level and reduce the use of heating and cooling, which have a high energy demand. Currently the majority of traditional insulation materials such as mineral wool are made from non-renewable resources. An alternative insulation material known as cellulose fibre insulation (CFI), has the benefit of using recycled paper to produce, while having similar performance properties of traditional insulation materials. Usually, cellulose insulation is blown dry in closed wall cavities or attics. A more recent method of installing CFI, known as the "wet spray" method, has the advantages of ensuring a proper distribution and filling of the material in a wall cavity, while preventing sagging through the use of pulverized water. The disadvantages associated with this method, however, are mainly due to improper installation of CFI while it is sprayed with water. If too much water is applied, or if its applied in a high humidity, low temperature conditions, the material could take a long time to dry, imposing a long delay in construction times, or worse promote mould growth within the material or surrounding wood structures. Too much sprayed water could also weigh down the material, making the material sag and causing voids in the installed wall cavity. One solution to these issues would be to include an additive that improves the drying and/or reinforces the material, reducing its initial water dosage. Ideally this additive should be based on natural or renewable resources in order to preserve the eco-friendly nature of the material. It is with this strategy in mind that the project of this thesis was conceived. The objectives of this project are to:

- Investigate the properties of cellulose fibre insulation
- Study the influence of water dosage on the performance characteristics of the material
- Characterise potential additives to be used with wet-spray CFI, study their influence on these characteristics and optimize the new formulation

- Model the dynamic hygrothermal performance of various formulations of CFI under different climatic conditions.

The work of this research project is collaboration from the Laboratoire de Chimie Agro-Industrielle (INP-ENSIACET) with French consulting firm Greenbuilding (the main coordinator of the project). It is financed in part through grants from the Association Nationale de Recherche et Technologie (ANRT) as well as Paris Région Entreprises. Material was kindly provided by SOPREMA.

The first chapter of this thesis is dedicated to the study the state of the art on cellulose insulation and the possible additives, biobased binders and surfactants which could improve its properties. The work in chapter two seeks to establish a foundation of the performance of cellulose insulation installed via the wet spray method, mainly the influence of the applied water dosage on its final properties. Chapter three focusses on the characterization and the screening of the potential additives, as well as their influence on the thermal and mechanical properties of cellulose insulation. Finally the last chapter further optimizes the material, details its performance under different conditions through hygrothermal modelling and, determines the properties of the final insulating material.

1

STATE OF THE ART

1.1 INTRODUCTION, CONTEXT AND OBJECTIVES

1.1.1 Sustainability in the building sector

One of the defining challenges confronting mankind in recent times is that of global warming. Manmade contributions to greenhouse gas emissions are the cause of the yearly increase in average worldwide temperatures (Pachauri et al., 2014). Numerous national and international initiatives are taking place in order to limit these effects, through the reduction of greenhouse gas emissions, such as, carbon dioxide (CO_2) (Stavins et al., 2014).

Energy efficiency in buildings is an important factor in contributing to the reduction of greenhouse gas emissions. The building and construction sector accounts for 30% to 40% of worldwide energy consumption, and (UNEP, 2007) with a large part belonging to the need to heat and cool buildings. It is with that in mind that many countries are looking to improve the energy efficiency of buildings, with directives such as the European directive 2010/31/EU, which states that new constructions in 2020 will have to consume 'nearly zero-energy' (European Parliament, 2010). One strategy for achieving this goal is through the optimal use and installation of materials that reduce the direct heating and cooling requirements to maintain thermal comfort in new and existing constructions. While these materials aid in reducing the *operational energy* consumption of buildings, most traditional insulation materials have a high *embodied energy* (Zabalza Bribián et al., 2011) which is defined as the total of the energy necessary for the entire

life cycle of a product. The lifecycle of a building material usually includes the stages of its production, installation, operation, maintenance and demolition. There is therefore a need to not only improve the energy footprint of buildings through improved insulation materials and standards but also to use alternative green materials with lower embodied energy.

1.1.2 Cellulose fibre insulation (CFI): a sustainable solution

The main role of thermal insulation materials in a building envelope is to prevent heat loss and provide thermal comfort for a building's interior. In order to do so, a material or composite of materials must provide a significant resistance to heat flow, and thus reduce a building's requirements for heating or cooling. The factor that characterizes an insulation material's effectiveness is its thermal conductivity λ (measured in W/mK). The lower a material's thermal conductivity, the more effective it is as an insulator, thus requiring a thinner layer to provide the same interior temperature.

It is defined as the amount of heat that can be through an area of unit thickness of material, within a defined temperature difference. The R value of a material is the reciprocal of thermal conductivity multiplied by its thickness (L/λ) and is used for calculating the thermal resistance to heat flow of any material.

Traditional insulation materials include glass fibre, stone wool, expanded polystyrene, and polyurethane foam. While these materials are efficient in maintaining thermal comfort to a building's interior, they are made with non-renewable resources and have a high embodied energy. Consequently, there is an increasing interest for alternative insulating materials that come from renewable or recycled fibres. Natural fibres such as jute, flax and hemp have shown to be suitable alternatives to mineral insulation and are the subject of numerous research projects (Madurwar et al., 2013).



Figure 1.1: Cellulose insulation fibres.

Cellulose fibre insulation, (CFI), a fibrous insulation material comprising of recycled paper fibre, can provide sufficient thermal comfort for a building's interior, while providing a more eco-friendly alternative to classical mineral based insulation materials. Despite these benefits, cellulose insulation remains an underused material. Despite recent trends in sustainable building initiatives, one study allocates the market share of cellulose insulation for new homes in the U.S. in 2013 to be under 10% (see Table 1.1), while both forms of fibreglass have a combined majority share of 75% (Home Innovation Research Labs, 2013). One cause for this lack of prevalence is due to the lack of knowledge on the performance, manufacture, and installation of CFI.

Shares of New U.S. Home Insulation Material		
	2012	2013
Fiberglass batt	54.0%	55.4%
Fiberglass blown	18.1%	19.1%
Rockwool	0.9%	1.1%
Cellulose	9.4%	9.1%
Spray foam	10.8%	8.3%
Foam board	5.6%	5.7%
Other	1.1%	1.3%
TOTAL	100.0%	100.0%

Table 1.1: Market share of US insulation materials in 2012 and 2013 source: (Home Innovation Research Labs, 2013)

This chapter will focus on the current state of the art of the topics of this project. Firstly, the major characteristics, properties and performance features of cellulose fibre insulation will be investigated. This will include its manufacture, installation and physical properties of both wet spray and dry cellulose insulation including the issue it presents with drying and opportunities in research to improve these properties. Then the feasible additives which could be used in conjunction with cellulose insulation to improve its properties are catalogued and detailed with regards to their properties and interactions with cellulose.

1.2 THE PROPERTIES OF CELLULOSE FIBRE INSULATION (CFI)

1.2.1 Composition

Cellulose fibre insulation is mainly composed of ground paper fibres treated with inorganic additives that act as fire retardants and mould growth inhibitors. Its consistency is similar to that of cotton wool. The source material for the cellulose fibres are usually recycled newspaper, coming from either unsold or recovered papers. Newsprint is generally manufactured by mechanical pulping. Recycled newsprint or chemical pulp could also be incorporated. As with most lignocellulosic fibres, newsprint is comprised of a mix of cellulose, hemicelluloses, and lignin. Unlike chemical pulping, mechanical pulping results in little removal of lignin content. Mineral and organic additives, such as kaolin, china clay or cationic starch are also incorporated into the paper pulp in order to improve such properties as paper opacity, moisture retention, and strength. The inks typically used in the paper are produced from inorganic carbons, with the chromatic inks coming from organic pigments. The average proportions of the main components in newsprint and office paper (chemical pulp) are presented in Table 1.2 (Wu et al., 2001). These amounts can vary according to different sources of paper with variable compositions and quality.

	cellulose %	hemicellulose %	lignin %	extractives %	proteins %	ash %
Office paper	67,4	13	0,93	0,7	0,31	11,6
Newsprint	48,3	18,1	22,1	1,6	0,44	2

Table 1.2: Average component proportions of newsprint and office paper, (Wu et al., 2001)

1.2.2 Production

As a final product, cellulose insulation can come in two forms: as a prefabricated panel, in which the cellulose fibres are moulded with polyester or a similar binder or, more commonly, the loose fibres are sold in bulk form to be manually applied on attics, ceilings, or walls. The first use of cellulose fibre as an insulation material can be traced back to 1919 in Canada (Siddiqui, 1989), but it was until the 1950s that commercial cellulose insulation products became commercially available in the US, where it was mostly used for attic retrofitting. CFI surged in popularity in the US in the 1970s due to an increased interest in energy performance following the American oil embargo of 1973. Nowadays cellulose insulation still used but it is eclipsed by mineral fibre insulation materials.

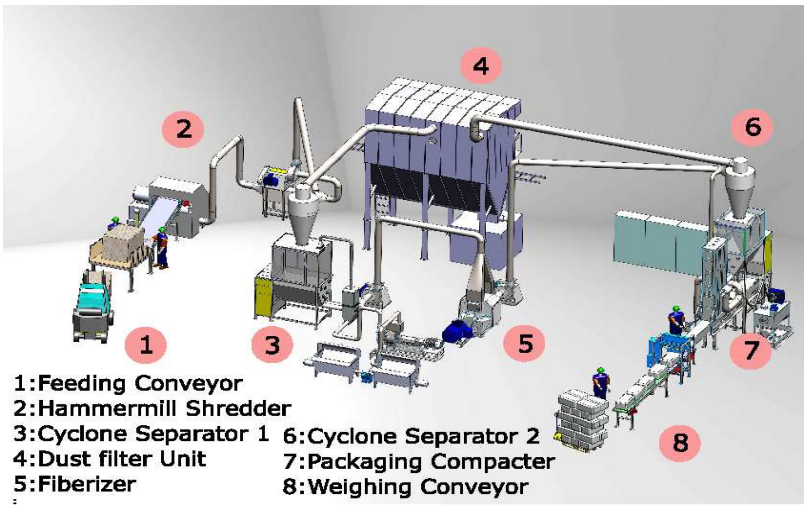


Figure 1.2: Example of a typical manufacturing process of CFI, adapted with permission from (Makron Engineering, n.d.)

In a typical production process of CFI (Figure 1.2), newspaper arrives in bulk to the manufacturer and is then sorted to remove any foreign objects. Items such as clips and plastics are removed, but also low quality or humid paper is also sorted. The newsprint

passes through a feeding conveyor (1) then is torn to smaller pieces that are between 2 and 4 cm in diameter in a shredder (2). The fibres then pass through a cyclone separator (3) in order to remove any remaining staples or other metallic elements. The fines from the shredded paper are blown through a filtering unit (4). The material then goes through a fiberizer (5) which uses high pressured air to reduce the paper into low-density cotton-like flakes, as shown in Figure 1.3. It is in this stage that the powdered additives are dispersed and mixed with the fibres. The additives used are typically a mix of borax and boric acid, with a dose of around 15%-20% of the mass of cellulose fibres. A second cyclone separator (6) then removes the fines created from the fiberizer. In the final stage of the process, the fibres are filled in bags and then mechanically compacted (7) into 3 times its normal density (around 130 kg/m^3), in order to reduce transport costs. The bags are then weighed (8) and bundled into pallets and transported to supplier or directly to construction sites.



Figure 1.3: Microscopic scan of cellulose fibres, 10X magnification

1.2.3 Installation of CFI

There exist two main methods of applying CFI. Depending on the desired properties, CFI can either be installed via the “loose fill” or the “wet spray” method.

In the loose fill method the cellulose fibres are installed with specific pneumatic blowing equipment. The compacted cellulose is fed to the blower which separates the fibres which then pass through the blowing system. The CFI is then delivered via air

pressure into closed wall/roof cavities or attics through a hose. When cellulose is installed as “dense pack”, sheets of netting are put in place over wall cavities. The cellulose is then blown into the cavities between studs at a higher density than loose fill, with the netting supporting the fibres. One of the disadvantages of this method is that settling of the material may occur over time, which decreases the insulation’s effectiveness, forming voids that cause thermal bridging in a building envelope (CIMA, 1998a).

The wet spray technique is mostly used in open-wall wood cavities separated by studs. It uses the same blowing equipment as with loose-fill CFI, but a separate pump is used to spray water simultaneously as the material is being blown with the cellulose in order to improve the adherence of the fibres. After projection the excess material is removed via a motorized wall scrubber and the excess moist material is reintegrated in the blower. The water/CFI mass ratio used in this process is typically around 40%-60%. Adhesives, either mixed with the water or dispersed within the fibres could also be used (CIMA, 1998b). The main disadvantage of this method is that drying times may vary, depending on the thickness and ambient conditions of installation. In some conditions the material may take months to dry out due to high humidity and low temperatures (Salonvaara et al., 2010). A variant of the wet spray method is known as “stabilized” cellulose where a smaller dosage of water (less than 20% in mass) is used to prevent dust and settling in horizontal applications.

1.2.4 Density and settling

Once cellulose insulation is installed, the loose fibres might compact and loose thickness over time. Therefore, when dealing with loose fibres as an insulating material, it is important to distinguish between the “blown” density and the “design” density of the fibres. The blown density is the declared density after installation in vertical or horizontal applications, and the design density (which takes settling into account) is determined via impact testing and/or cyclic humidity testing. Impact testing consists of subjecting the loose cellulose samples to a series of vibrations. In cyclic humidity tests, the samples are subjected to periodic variations of relative humidity (AFNOR, 2014). One of the first studies regarding the settling of CFI was done by Bomberg and Shirtliffe (1979). Their study found an average blown density of 34.8 kg/m^3 for horizontal applications. The

average loss in thickness from settling was 21.5% wherein 10.5% was from drop impact tests and 11% was from cyclic humidity testing. The design density can be then calculated using by multiplying a factor which takes into account both types of settling:

$$D_d = (100/(100-S))\ D_i \quad (1.1)$$

Where D_d is the design density, D_i is the installed density, and S is the sum of both the settling from drop impact tests and cyclic humidity testing.

The previous values give a design density factor of $1.27D_i$, thus an average design density of 44.4 kg/m^3 for horizontal applications. It was also found that the dosage of fire additives increases density linearly, although the type of additive or mix thereof has little influence on final density. A survey of 38 houses in six Canadian cities (Zaborniak, 1989) found the actual settling density, a year after installation, to be averaged to 11.1%, with a range of 8.3%. The study suggests that the blown density measured in laboratory be first multiplied by a factor of 1.074 to account for differences between lab and building site measurements, and then calculated with equation 1.1 using an average settling of 11.1%.

For horizontal applications the compressibility of loose-fill CFI can make its density vary widely. One early study by Bomberg and Solvason, (1980), shows installed density varying between 50 to 90 kg/m^3. It was recommended to increase density by 10% after filling the wall cavity in order to prevent settling, with a minimum density of 57 kg/m3. A series of works by Rasmussen (2005, 2003, 2002, 2001) have produced an approach which allows to analytically determine the optimal installed density of loose fill CFI that prevents settling in wood frame walls. The method takes into account the dynamic mechanical behaviour of a typical insulated wall cavity that is subjected to a cyclical variation in humidity in order to determine the density required for the fibres to lose volume. The volume stable density of CFI was determined through the study of the creep, coefficient of friction, and horizontal stress ratio testing of loose fibres. As an example, the minimum density to prevent settling with CFI a 2.4m tall, 0.1m thick and 1m wide gypsum wall at 25°C and 50% RH was found to be 48 kg/m^3. This value increases linearly with wall thickness and relative humidity (Figure 1.4). Dynamic conditions were also tested, where humidity varied from 50% to 80%. In this case a 2.3m high, 0.198m thick and 0.495m wide gypsum board cavity was calculated to require a

density of 62.3 kg/m^3 to prevent settling. This was later confirmed experimentally with a CFI-filled cavity with a density of 62.7 kg/m^3 where settling was not observed.

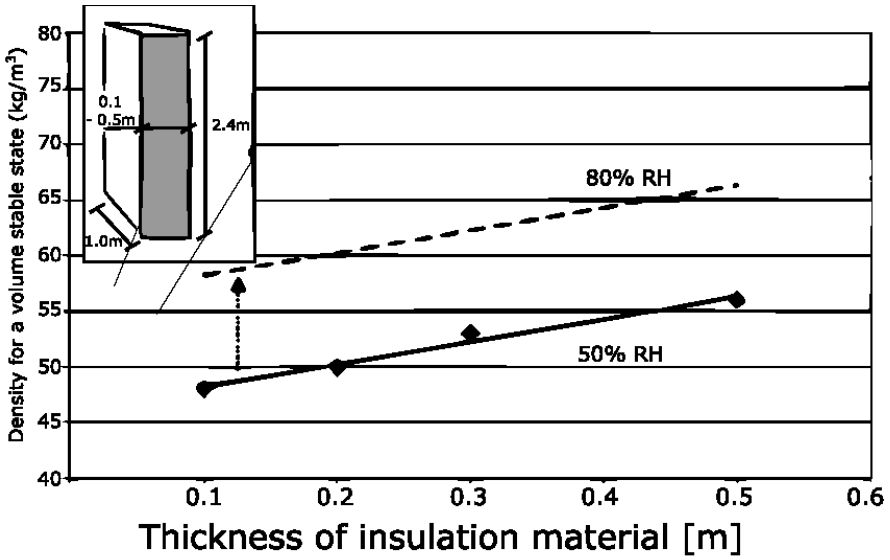


Figure 1.4: Calculated minimal density for settling prevention of loose fill CFI in a wall under static humidity conditions (50% and 80% RH), as a function installed thickness, top left shows the dimensions of the wall cavity (Rasmussen, 2002)

For wet spray applications, the dry density of CFI has been shown to increase linearly with installed moisture content, ranging from 39.6 kg/m^3 with 40% moisture content to 71.3 kg/m^3 with 100% moisture content (Salonvaara et al., 2010). If installed properly, wet spray cellulose does not settle. For stabilized cellulose, an initial moist density of around 45 kg/m^3 gets reduced to around 38 kg/m^3 after drying. Settling with the stabilized cellulose method in attics was found to be reduced to around 5% (Graves and Yarbrough, 1989).

1.2.5 Thermal properties

Although the typical value for CFI's thermal conductivity is around 0,040 W/mK, its properties and performance can vary slightly depending on manufacturing and method of installation. The work of Kwon and Yarbrough (2004) has shown that a difference in the source newsprint quality can affect thermal performance. In their study, CFI samples coming from US and Korea were measured through heat flow meters, in accordance with ASTM C 518. By comparing CFI from both countries, the study found that the Korean

fibres that are shorter due to having gone through more recycling processes show a higher value for thermal conductivity, and therefore lower insulating performance than CFI fibres from the US.

Since cellulose fibres are naturally hygroscopic, moisture absorption can also affect thermal conductivity values. Tye and Spinney (1979) studied loose fill CFI installed in ceiling and wall constructions subjected to cyclic thermal and moisture gradients. Thermal conductivity measurements were made on installed samples using the standard ASTM C236 guarded hot box method with a mean temperature of 15°C and a temperature difference of approximately 10°C. It was found that thermal conductivity increased by 15% for a moisture gain of 10%. Nicolajsen (2005) found that under the hygroscopic range (i.e. RH <90%) the change in thermal transmittance of loose fill cellulose insulation within a wall cavity was not significant (1% to 3% increase). The study was done on facade elements with 285mm loose-fill CFI equipped with heat flow meters and moisture measuring dowels. Heat flow measurements were made according to the DS 418 standard. Sandberg (1992) developed three approaches to determine thermal conductivity as a function of water absorption using moisture content profiles of cellulose insulation. Measurements were made on 164mm thick loose fill CFI samples on 600mmx600mm frames, following the ISO 8301 and ISO DIS10-051 standards. Computer simulations used the following relation with regards to the thermal conductivity of cellulose:

$$\lambda=0.037 + 0.0002' w \ (W/mK) \quad (1.2)$$

Where w is the mass of water per unit volume of cellulose kg/m^3. The calculated results were in agreement with sample measurements.

Talukdar et al. (2007) determined a polynomial function to describe the relation between moisture and thermal conductivity by curve fitting values measured by a heat flow meter apparatus according to ASTM standard C518 on cellulose at different relative humidity conditions. Measurement temperatures were at 10 °C and 35 °C, with an average temperature of 22.5 °C

$$\lambda_{eff} = (a+b\varphi+c\varphi^{1.5} + d\ exp(-\varphi)) \quad (1.3)$$

Where a = 0.092482655, b = 0.15480621, c = 0.066517733 and d = 0.1296168.

The only research that studied changes in thermal conductivity past the hygroscopic range of 90% RH was done by Vèjelis et al. (2006). Their study determined moisture content of CFI in one and two floor buildings with masonry walls with different thickness of insulation throughout various moisture periods measurements. A qualitative method was used to determine the influence of moisture on variations in thermal conductivity. An increase in 1% of moisture content can lead to an average increase of 1.2 % to 1.5 % in λ values for loose-fill CFI. Even when high moisture content was reached, thermal conductivity increased from 1.6 - 2.0 % for 1 % of moisture content (Figure 1.5). These changes in values of λ are similar to those mentioned previously by Nicolajsen (2005) and Talukdar et al., (2007) in the hygroscopic range. Generally for the hygroscopic range, the increase in thermal conductivity could be considered negligible. It is only when capillary moisture begins (RH > 90%) that the insulating properties start to decrease significantly. Such cases could arrive due to rain infiltration, leaking pipes, or improperly installed wet spray cellulose.

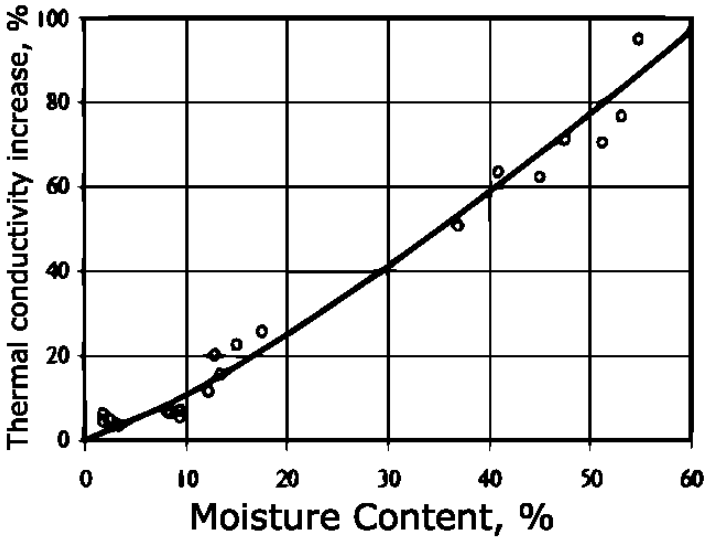


Figure 1.5: Increase in thermal conductivity with moisture content of cellulose fibre insulation (Véjelis et al., 2006)

1.2.6 Air infiltration

Openings in a building's envelope can cause air leakage which can have an impact on energy loads in buildings. The most cited study by manufacturers regarding air infiltration with CFI was done by Boonyakarn et al (1990). The study found that the installation of CFI (wet-spray in walls and loose-fill in attics) reduced the air change rate from 87.5 to 29.4 ACH (air changes per hour) with 50 Pa blower door testing. CFI was found to improve air tightness 36% better than fibreglass.

A study by the North American Insulation Manufacturers Association (NAHB Research Center, 2009) found that, via testing in wood framed walls, wet spray cellulose had reduced the air infiltration of a structure from 10.5 to 2.2 m^3/h (converted from cubic feet per minute) while fibreglass batts only reduced it to 6.1 m^3/h . The difference in air infiltration between types of insulation is negligible once a weather barrier was applied.

1.2.7 Fire properties:

The high flammability of cellulosic fibres requires them to be treated before installation in order to achieve acceptable levels of combustion and smouldering resistance. In a typical CFI material, borate salts are added to prevent combustion and boric acid is added to prevent smouldering (Sprague and Shen, 1979). Other additives include: aluminium sulphate, aluminium trihydrate, ammonium phosphate, and ammonium sulphate. Day and Wiles (1978) studied the influence of the proportions of these additives on flame spread and smouldering resistance. The minimum boric acid required to prevent smouldering as a function of borax dosage was established:

Boric acid required = 11.6 + 0.185 x (borax used).

(1.4)

Day et al.(1980) found that the optimal borax/boric acid ratio of 1/8 with a dosage 16% is necessary to prevent both flaming and smouldering combustion (Figure 1.6).

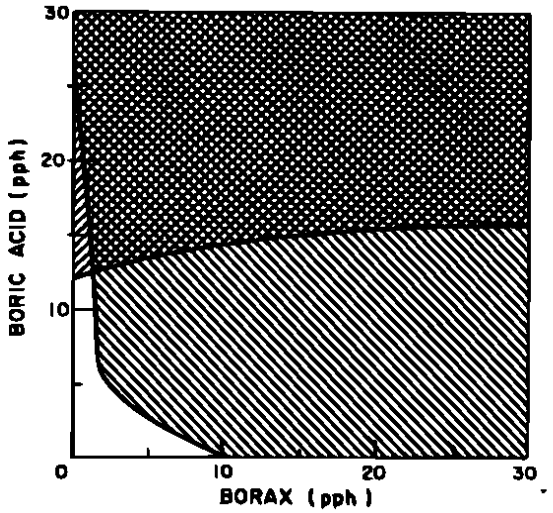


Figure 1.6: Proportions (parts per hundred) of borax and boric acid in order to achieve smoulder retardancy (// diagonal) and flame retardancy (\\ diagonal) (Day et al., 1980).

A three component formulation using borax, boric acid, and aluminium sulphate was also studied. Varying dosage from 12%, 18%, and 24% increases the possible proportions of these constituents which allow both smouldering and combustion resistance to be obtained. In another study (Day et al., 1981), the effect of wetting on additives was examined. They establish that wetting and drying of the CFI caused a higher concentration of both borax and boric acid to appear on the surface of the material. This migration did not affect smouldering resistance and would actually be favourable for flame combustion resistance. Sprague (1979) studied the consistency of formulations and found variability in the distribution of test results. Samples were found to attain class I or II flame resistance with a variable distribution. As additive dosage increased, this variability was reduced. Some of the variability was due to inconsistency in the testing method itself.

1.2.8 Fungal development

It is widely known that wet lignocellulosic materials can allow mould growth. In the case of CFI, the added additives can serve a dual purpose of preventing mould growth as well as fire propagation. In the work of Herrera (2005), it was found that the boron included in the cellulose was found to have a sporocidal effect on five of the most common types of fungal spores, even when subjected to a high concentration of fungi.

For untreated fibres exposed to fungal samples, moisture content and relative humidity was found to have an influence on the fungal growth rate of cellulose insulation. As the CFI samples dried, the rate of mould growth decreased.

There exist however, case studies where mould growth has been found to be produced in houses insulated with CFI. Godish and Godish (2006) studied four wet spray CFI-insulated houses where mould was prevalent. While the conditions in which the wet spray CFI was applied were not detailed (i.e. high water dosage), it was found that two of the houses developed fungi due to rewetting of the fibres because of water infiltration. Numerous hydrophilic xenophilic and toxigenic species of fungus were found both within the CFI material and in airborne samples. While this mould exposure poses a risk to building occupants, properly applied wet-spray CFI should not present these problems.

1.2.9 Life Cycle Analysis

As mentioned before, CFI has a low embodied energy compared to traditional mineral and natural insulation materials. A comparative analysis with three impact categories of the life cycle analysis (LCA) of common insulation materials was presented by Zabalza Bribián et al. (2011). It is worth noting that the functional unit is 1 kg of material. Since the materials have different densities and thermal conductivities, a more proper functional unit would be the necessary amount of material to provide a specific value of thermal resistance.

	Building product Density (kg/m^3)	Thermal conductivity (W/mK)	Primary energy demand (MJ-Eq/kg)	Global Warming Potential (kg CO_2-Eq/kg)	Water demand (l/kg)
EPS foam slab	30	0.0375	105.486	7.336	192.729
Rock wool	60	0.04	26.393	1.511	32.384
Polyurethane rigid foam	30	0.032	103.782	6.788	350.982
Cork slab	150	0.049	51.517	0.807	30.337
Cellulose fibre	50	0.04	10.487	1.831	20.789
Wood wool	180	0.07	20.267	0.124	2.763

Table 1.3: Comparative life cycle analysis of common building materials (Zabalza Bribián et al., 2011)

A more in depth LCA comparison was done by Schmidt et al. (2004), who studied the cradle to grave assessment of stone wool, flax, and CFI, in compliance with the LCA standard ISO 14040. In this case the functional unit was the amount of material necessary

to provide a thermal resistance of 1 m²K/W so 1.280 kg of material in the case of loose fill CFI. The study takes into account the production of newsprint, the manufacture of CFI, the incorporation of its additives, its installation, use and disposal for the calculation of its inventory. Sensitivity analysis of the end of life stage was also considered. For loose fill solutions, manufacturers state that CFI can be recycled if no contaminants are present (Boogman, 2014), or incinerated to provide energy in a waste incineration plant. The study analyses the impact of partial recycling incineration or landfilling of the material. The highest impact was caused by partial landfilling of the CFI, which nearly tripled the global warming impact factor, due to the amount of methane released by the material. Recycling vs incineration had less of an impact. Interestingly, in this study CFI showed a higher total energy consumption than stone wool (for the same functional unit), which contradicts the studies previously shown. One reason for this could be strategy involved in the consideration of the impact of the manufacture of newsprint. In this study, newsprint production represented over 90% of overall energy consumption in the LCA of CFI. This highlights the importance of the initial hypotheses when analysing the life cycle of a recycled material such as CFI. More recently, a review on thermal insulation materials done by Schiavoni et al. (2016), showed cellulose to have one of the lowest embodied energy and global warming potential among most common insulation materials.

Life cycle assessment is a useful tool in material selection for full construction projects. Takano et al. (2014) studied the impact of building material selection on the environmental characteristics of a construction in Finland. It was found that the change from rock wool to CFI as an insulator could reduce embodied energy of the building envelope by 15%. Similarly Tettey et al. (2014) studied the influence of different insulation materials on primary energy and CO_2 emission of a residential multi-storey building using the criteria of BBR 2012 and Passivhaus 2012 energy-efficiency standards. It was found that the replacement of stone wool by CFI on most parts of the building envelope resulted in global energy reduction of 6% to 7% and a decrease in CO2 emissions from material production of 6% to 8%, depending on the standard chosen (BBR 2012 or Passivhaaus). This is an interesting factor to consider when dealing with the refurbishment of buildings, where not only the replacement of old mineral insulation

with CFI will improve thermal properties of a refurbished building, but also reduce its ecological impact.

1.2.10 Moisture properties

The behaviour of a building material with moisture can be determined by a series of intrinsic parameters. The sorption isotherm of a material can determine amount of water absorbed under different values of relative humidity. This series of values is usually measured through continuous weighing of a cellulose insulation sample subjected a series of changes in humidity via saturated salt solutions.

Sorption and desorption isotherms were determined experimentally by Hansen et al. (2001) (Figure 1.7). The isotherms are measured at 20.0ºC ± 0.5°C in a test chamber as described in EN ISO 12571, a magnesium perchlorate solution was used as a desiccant. The difference between sorption and desorption values (hysteresis) was negligible. Untreated CFI had a slightly lower sorption curve than treated CFI, suggesting that the mineral additives contribute to the adsorption of ambient humidity. A similar sorption curve was found by Talukdar et al. (2007).

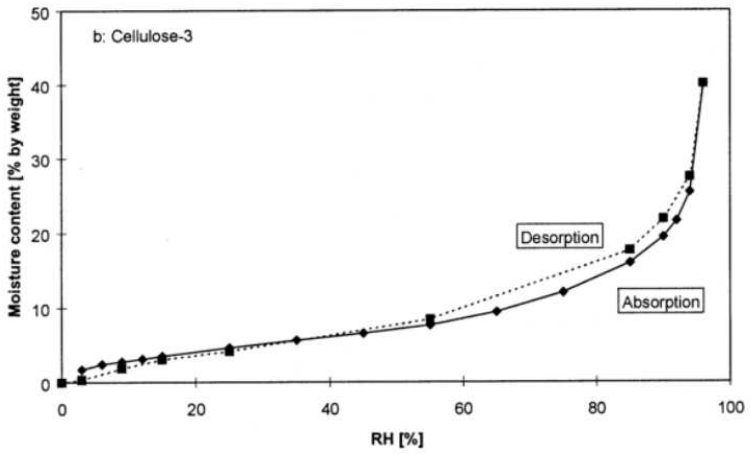


Figure 1.7: Sorption-desorption isotherm of treated cellulose insulation

Moisture diffusivity is a property that is used in simulations to determine the moisture concentration profile of a material. It is defined by the moisture transport equation:

$$J_m = -\rho . D . grad\ u \quad (1.4)$$

With J_m= moisture flux (kg/m²s), ρ the dry density of the material (kg/m3), and u the moisture content [(kg/kg)]. This parameter was determined by Marchand and Kumaran (1994). Samples of blown CFI were subjected to moisture intake and then continuously scanned via gamma ray attenuation. These scans provided the moisture content profiles within the material as a function of time. Through Boltzmann transformation of these profiles, the moisture diffusivity D was determined as a function of moisture content within the cellulose. The value of D varied exponentially from $5x10^{-8}$ m/s² to $1.2x10^{-7}$ m/s² for moisture contents of approximately 10% to 175%.

The water vapour permeability is the rate in which water vapour is transported through materials. This characteristic defines the "breathability" of a material. Hansen et al. (2001) determined the value of vapour permeability of CFI from cup measurements at 23ºC varying from 50% to 94% RH according to EN ISO 12572: 177±29 x10 $^{-12}$ kg/(Pa.m.s). An increase in density (from 40 to 65 kg/m^3) greatly reduced the permeability, while the removal of mineral additives had less of an impact. The values are similar to those found in other works: (Mortensen et al., 2005), (Peuhkuri et al., 2008), and (Kalamees and Vinha, 2003). Talukdar et al (2007) established the water vapour permeability of CFI as a function of relative humidity using ASTM Standard E96/E96M-05.

A parameter that is frequently cited by manufacturers is the moisture buffering value (MBV) which is the ability of the materials within the room to moderate variations in the relative humidity. Cerolini et al.(2009) calculated the MBV of CFI by exposing 69.6 g of CFI to daily cyclic exposure of high (75%) and low (33%) relative humidity levels for 8 h and 16 h. The moisture buffering value of CFI was found to be around 3.06 g/m².%RH, which can be classified as an "excellent" moisture buffer according to the scale established by Rode et al. (2007).

The highly hygroscopic nature of cellulose insulation can be detrimental to CFI's performance, as was shown with the two previous sections. However having a hygroscopic material in a building envelope could theoretically be beneficial when it comes to regulating humidity conditions inside a building, especially if a vapour retarder

is not integrated in the building envelope. Rode (1998) modelled the performance of a CFI wall under isothermal and nonisothermal conditions in Nordic climate. In the case where no moisture barrier or plasterboard was applied a small improvement in interior relative humidity was found for winter months. However, external humidity conditions caused moisture accumulation within the CFI to reach levels over 90% RH, which could potentially promote mould growth. Hagentoft and Harderup (1996) used hygrothermal 1D models to calculate moisture uptake of a typical wall with a brick façade and thermal loose fill CFI insulation exposed to Swedish climate. The study found that in when vapour retarder is not used, moisture accumulation can reach critical levels and possibly cause mould growth in the wooden elements of the wall. In the work of Vrána and Gudmundsson (2010), moisture transport within CFI was measured experimentally in order to model its behaviour under massive condensation and sub-zero temperatures that create ice formation. Their studies found that ice formation had little influence on the water vapour permeability of the material, yet the material continued to accumulate moisture and did not reach a steady state within the testing period of 100 hours.

Using a full scale testing chamber subjected to moisture load, Mortensen et al. (2005) found that CFI can reduce interior relative humidity peaks by up to one half, but as with the previous studies, this moisture reduction becomes negligible once the surface layers of the composite wall are covered in plasterboard.

This moisture sensitivity accentuates the need to protect the material from moisture infiltration, through the use of external weather barriers.

1.2.11 Drying of wet spray CFI

For wet spray cellulose, drying is an important factor to consider during installation. The water from the sprayed fibres could be transmitted to wood frames cavities which could cause warping or mould growth. A study by the Canada Mortgage and Housing Corporation (CMHC, 1990) found that the cellulose increased plywood sheathing moisture content to 24% 30 days after installation, which then reduced to 15% after 260 days. The critical moisture content in which wood starts to develop fungi is around 30% so the moisture values were below the threshold although for different conditions, higher moisture is possible. Salonvaara et al (2010) studied drying of wet

spray CFI with a hygrothermal model that takes into account the period of installation. The study compared drying in a region with warm dry climate vs. a region with cold humid climate. It was found that for winter months the wet spray CFI would take many weeks to dry and in some cases never dry at all, especially in colder region. For example Figure 1.8 shows that, during the winter months of November, December, and January (solid blue, dashed orange, and solid black lines in the graph, respectively) moisture content decreased by only 10% in a month. The wet spray method is therefore preferred to be applied in warmer drier climates.

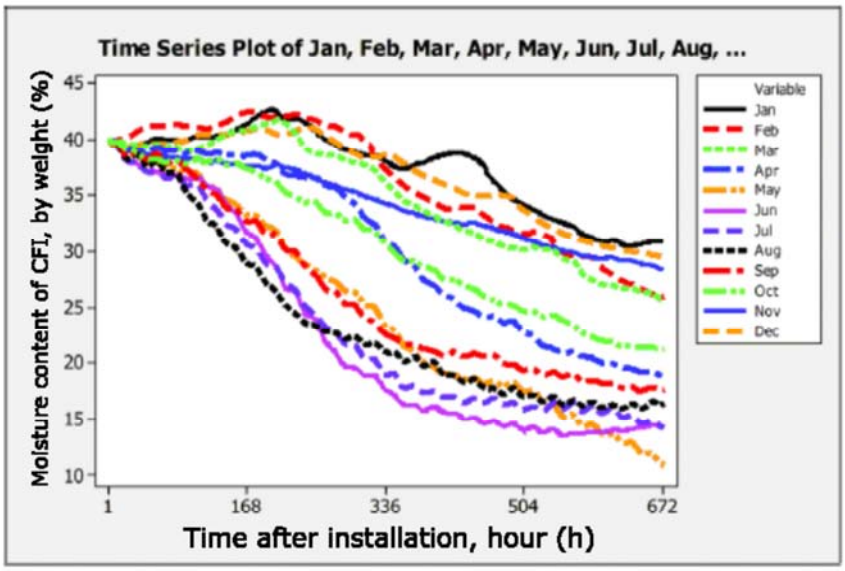


Figure 1.8: Calculated evolution of average moisture content in exterior facing half of wet spray CFI with varying months of installation in Detroit Michigan (Salonvaara et al., 2010)

1.2.12 Present opportunities in cellulose insulation research

The state of the art of cellulose insulation has shown both its benefits and disadvantages. While its physical properties have been thoroughly studied especially with regards to the dry blown method, the wet sprayed technique is underrepresented in the literature. Firstly, no work has been made to characterize the mechanical performance of the material after it is sprayed. The influence of the sprayed dosage on its properties has not been presented. Finally no research has been dedicated to improve the problem with regards to drying. As of now, only water has been used with the wet spray method, and

additives have not yet been considered. The use of additives could improve the mechanical cohesion of the material, and/or improve its drying times. The main objective of this work is then to further characterize cellulose insulation installed via the wet spray method, and to search for a feasible additive which could improve its drying and mechanical properties, while maintaining its positive qualities as an insulating material.

1.3 BIOBASED ADDITIVES AND INTERACTIONS WITH CELLULOSE

1.3.1 Adhesives: an introduction

An adhesive can be defined as a substance or group of substances that, once applied to a surface, can bind with that surface and resist separation or breaking. In doing so, an adhesive allows the uniform transmission of stresses from one surface to another. Some of the first uses of adhesives can be traced back to 4000 B.C, where prehistoric pottery was found to be repaired using tree sap. Another notable recorded use of adhesives can be traced back to ancient Egypt, in the period between 1500-1000 B.C where there existed paintings and murals portraying wood gluing operations. It was found that for the construction of king Tutankhamun's tomb, animal glue was employed to seal the casket (Nicholson et al., 1991; Rifai and El Hadidi, 2010).There are many examples throughout history of the use of animal binders such as casein, gelatine and blood glues. As technology progressed, the composition and performance of binders have evolved. The past century introduced adhesives using petroleum based synthetic polymers. In the present day, adhesives are employed for a variety of uses. New developments in formulations of adhesives are continuously being created to suit the required properties of different materials, for use in many industrial sectors such as: aerospace, automotive, construction, cosmetics, paper, packaging, food, and agriculture, amongst others. The most prevalent adhesives used today are based on polymers sourced from fossil fuels, such as urea-formaldehyde, epoxy, polyurethane, and vinylic and acrylic polymers. There is a current predisposition in the adhesive industry to return to the use of natural binders from animal and plant origin. Manufacturers seek alternative natural and renewable resources for which adhesives could be produced. This is mainly due to the decrease in

reserves of fossil fuels, their increase in price, and their negative impact on the environment and human health. There exist numerous biobased polymers which offer a sustainable alternative to traditional adhesives, while providing similar properties in terms of mechanical resistance (Imam et al., 2013) .

The ways in which adhesives could contribute to the drying of wet spray cellulose insulation are numerous. First of all, the hardening of the binder after it is sprayed with cellulose makes it consume the water that needs to be evaporated from the material, thus accelerating drying. The fact that the adhesive could reinforce cellulose insulation could make it that less initial water dosage is required. Finally the increased initial solids content from the adhesive makes the initial moisture content decrease, thus reducing the time required to reach the dry state. Ideally the included binder should not hinder the low environmental impact of the final product so this study focusses primarily on biobased adhesives.

1.3.2 Mechanisms of adhesion

A number of theories on the mechanisms that characterise the bonding of adhesives with their substrate have been developed. No one theory can describe all the interactions that take place once a material adheres to another. It is generally accepted that there is a combination of these mechanisms working together which leads to adhesion (Fourche, 1995). In recent years, it has been proposed that some of the classical theories of adhesion are a component of more complete models (Ebnesajjad, 2008). The current proposed theories are as follows:

Adsorption theory:

Reactions at the surfaces of both the adhesive and the substrate at the time they are in contact, cause specific interatomic and intermolecular forces. Adsorption theory encompasses all type of bond created from these forces, whether it be a primary bond, such as ionic, covalent or metallic, or secondary, such as Hydrogen or Van der Waals bonds. The nature of the bond depends on the properties of both the adhesive and the substrate. Secondary bonds present much less energy than primary bonds. Table 1.4 shows the typical bond energies of different types of bonds (Pizzi, 1994).

Type	Bond energy (kJ mol^{-1})
Primary bonds	
Ionic	600–1100
Covalent	60–700
Metallic, coordination	110–350
Donor–acceptor bonds	
Brønsted acid–base interactions (i.e., up to a primary ionic bond)	Up to 1000
Lewis acid–base interactions	Up to 80
Secondary bonds	
Hydrogen bonds (excluding fluorine)	1–25
Van der Waals bonds	
Permanent dipole–dipole interactions	4–20
Dipole-induced dipole interactions	Less than 2
Dispersion (London) forces	0.08–40

Table 1.4: bond energies of primary and secondary bonds (Pizzi, 1994)

Some works differentiate the adhesion theory with the type of bond, such as primary bonds are sometimes referred to as chemical adhesion (Kendall, 1994), (Ebnesajjad, 2008), and covalent adhesion is referenced when referring to covalent bonds in wood adhesives (Pizzi, 1994).

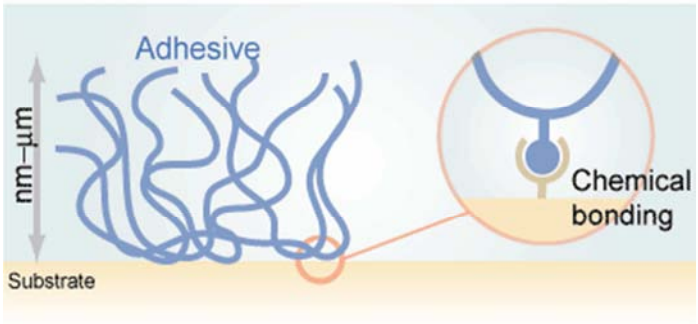


Figure 1.9: Adsorption bonding adhesion (Mattson, 2010).

Mechanical interlocking theory:

This is also known as the “hammer and nail” mechanism. Once the fluid adhesive flows through the substrate, it hardens and interlocks within the material, and thus increasing strength. This mechanism is especially prevalent when the substrate is porous, such as in fabrics, paper, and natural fibres, where a liquid adhesive can penetrate into the material by capillarity.

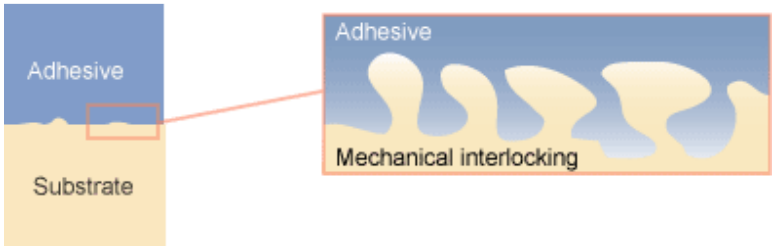


Figure 1.10: Mechanical interlocking adhesion (Mattson, 2010)

Diffusion theory:

Interactions between polymer chains at their interface of some materials induce adhesion. The entangling of polymer chains is affected by contact time, temperature, chemical properties, and physical form. The phenomenon only occurs with polymers with long molecules that allow movement and entanglement with each other.

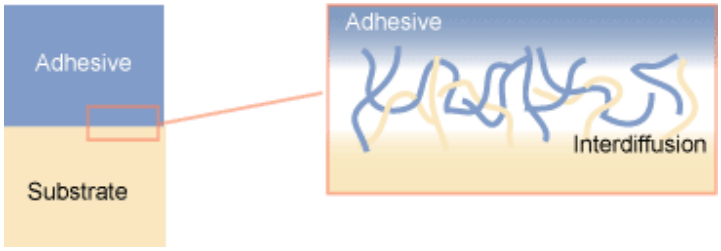


Figure 1.11: adsorption bonding illustration (Mattson, 2010)

Electrostatic forces theory:

Another factor in bonding between an adhesive to its substrate could be electrostatic forces. These forces occur when an electrical double layer of separated charges at the interface is formed and are believed to play a role in the resistance to separation of adhered materials. In order for electrostatic adhesion to occur, both materials must show properties that allow electron transfer at the interface. Adhesives and substrates that contain polar molecules or permanent dipoles are most likely to form electrostatic bonds. This type of adhesion mechanism is quite weak compared to the ones previously cited.

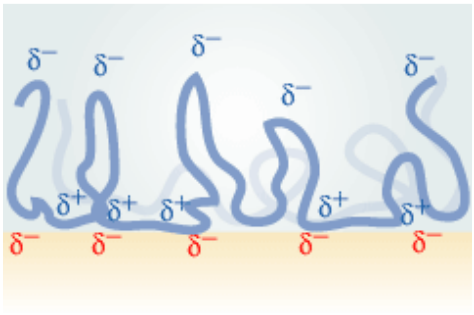


Figure 1.12: Electrostatic adhesion (Mattson, 2010).

Weak boundary layer theory:

This model is used to describe adhesive failure that occurs at the interface. One example of weak boundary layers is when air voids are formed through improper contact at the interface. This can occur through improper wetting of the adhesive. Another cause could be contaminants or impurities present in the interface, causing unfavourable reactions and voids between substrate and adhesive. In order to limit the effects of weak boundary layers, proper surface treatments surface treatments on the substrate can be applied so a strong adhesive bond can be achieved.

1.3.3 Adhesion in cellulose fibres

The closest available analogue to the adhesion of the moistened fibres from wet spray cellulose insulation would have to be the bonding of cellulose during paper manufacture. This association comes with two important distinctions however: In the paper manufacturing process: much more water is used than for the wet spray method of applying CFI (paper pulp is usually a slurry with low solids content, compared to the low moisture content of CFI), and the source material is slightly different (most paper pulp is refined and has been chemically treated to remove lignin and impurities, whereas CFI fibres are more representative of an unrefined, untreated mechanical recycled paper pulp) (Biermann, 1993). Nevertheless the adhesion process during paper manufacture can give an idea of the interactions of cellulose fibre insulation with water and adhesives. The primary mechanism of adhesion between cellulose fibres is generally accepted to be hydrogen bonding. Since cellulose is composed of hydroxyl groups, the oxygen atoms from these groups can form hydrogen bonds with the water molecules in contact with the fibres. As the paper pulp is pressed and dries these bonds between water and fibres are

replaced with interfibre bonds. However, the roughness of paper pulp (ranging from 10 to 10000 nm) can limit the surface in which cellulose microfibrils could be in contact in order to create hydrogen bonds, which are generally formed at a contact distance less than 1 nm (Fornué et al., 2011). This means that other adhesion mechanisms must also take place in order for the cellulose fibres to come in close contact and bond. Of the mechanisms mentioned beforehand, there are three main mechanisms that have currently been proposed as being dominant when describing the adhesion of cellulosic fibres: adsorption/chemical interactions such as hydrogen bonding and Van Der Waals forces, intermolecular diffusion between molecular chains, and mechanical interlocking of irregular surfaces (Gardner et al., 2008). The interdifusion theory posed by Voiutskii (1963) equates the cellulose fibre surface with a polyelectrolyte gel. During pressing and drying of the fibres, molecules diffuse into each other through inter-solubility, consequently close enough contact is made between molecules that hydrogen bonding is possible. Van der Waals forces have also been said to contribute to the adhesion of paper. These forces are created as a result of temporary dipoles from the movement of electrical atoms in molecules. The charged fibre surfaces in water create a double layer force, increasing surface adhesion. These adhesive forces depend on the charge density at the surface of the fibres as well as properties of the aqueous medium, such as pH and salt concentration (Eriksson, 2006; Yan and Li, 2012). Mechanical bonding could be applied when cellulose fibres and microfibrils interlock with each other. This mechanism is more obvious when dealing with adhesion of dried fibres, and is considered less impactful to overall adhesive strength of wet paper pulp. Incidentally this mechanism could explain the cohesion obtained with loose fill (dry) cellulose fibre insulation.

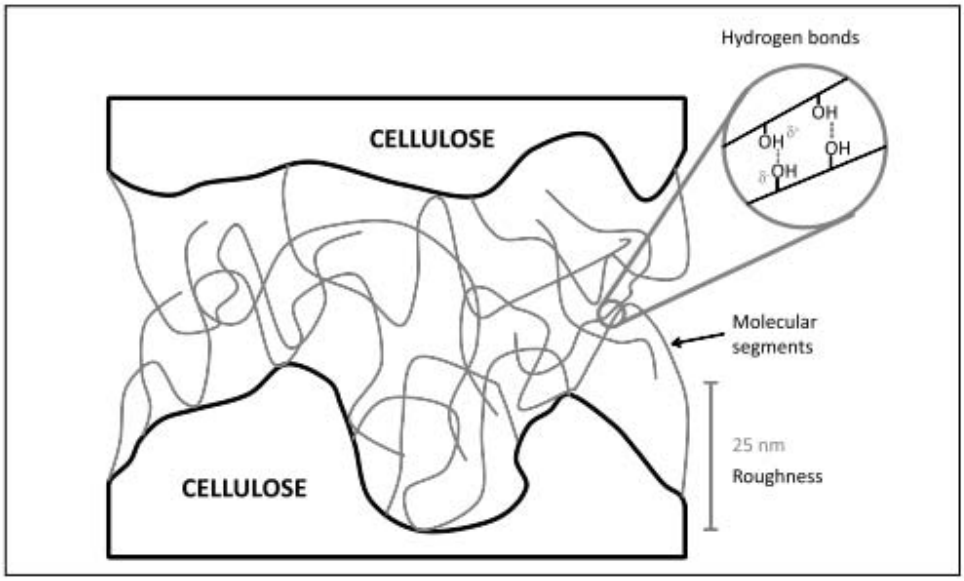


Figure 1.13: Schematic representation of microfibril formation of hydrogen bonds between cellulosic fibres (Gardner et al., 2008).

1.3.4 Types of adhesives in natural fibre composites

Adhesives and binders used in fibrous composites could also be defined through the reactions that allow them to flow, polymerize and harden. Most adhesives are in a liquid form which, once applied, "wet" the substrate and flows through its structure. This is usually achieved via either: (1) the use of heat, which allows the material to flow, (2) dissolving the material in a solvent, or (3) through polymerization of the liquid monomer form. Once in place, after the adhesive has flowed through the substrate, the material must cure in order to achieve its final form and reinforce the substrate. Most adhesive harden either through: cooling, evaporation of the solvent or chemical reaction. These reactions during the wetting flowing and curing stages define how they can be applied with a substrate (Ebnesajjad, 2008; Pizzi and Mittal, 2003). The main types include:

Thermoset: With thermoset adhesives, the application of heat induces polymerization and cross-linking of the adhesive, such examples include: Urea formaldehyde, phenol formaldehyde, acrylics and epoxies.

Thermoplastic: In the case of thermoplastic adhesives, heat is also employed but to is applied to either melt (semi-crystalline) or raise the temperature above the glass transition temperature (amorphous) thus allowing a polymer to flow through its substrate.

Ethylene Vinyl Acetate and Poly Lactic Acid are some examples of adhesives that portray these properties.

Solvent loss: The mechanism which characterizes solvent loss adhesives occurs when a polymer is dispersed or dissolved in a solvent, forming a solution in which over time, the solvent evaporates leaving only solid film. Glues such as polyvinyl acetate (PVA) – and starch are some examples of solvent loss adhesives.

Reactive systems: Adhesion with this type of system is realized through the mixing of two or more components that react together and form a chemically crosslinked bond, usually without using heat. They are known for providing high strength bonds and good performance. The most common reactive adhesives are epoxies, where a base resin and a hardener are mixed and can cure and harden at room temperature.

Others: There exist other, less prominent types of adhesives which activate or cure through different means. Some examples include moisture-curing adhesives such as some types of polyurethane which cure in when in contact with moisture, or anaerobic adhesives such as dimethyl acrylate ester, which flow when in presence of oxygen but harden in its absence.

1.3.5 Biobased adhesives

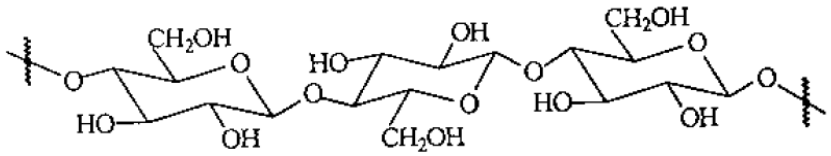
The main adhesives and gums coming from animal and plant resources are mainly polysaccharides, proteins, or phenolics, and can be classified from their main component as follows:

1.3.5.1 Cellulose esters

Cellulose is one of the most abundant polymers in nature. It is the main component of most trees, plants, crops and biomass, providing structure to their cell walls. Its structure consists of linear *D*-glucose units linked by β-(1→4)-glycosidic bonds (Figure 1.14).

Cellulose is mainly used in the manufacture of pulp and paper, but can also be applied to the production of biocomposites as a reinforcing fibre. As a binder, cellulose derivatives known are cellulose esters can contain adhesive properties or be used as an additive in adhesive, coatings, and paint formulations. Some examples of cellulose esters include: methylcellulose (MC), hydroxypropylmethylcellulose (HPMC), and

carboxymethylcellulose (CMC) (Nasatto et al., 2015). These cellulose derivatives have many uses as thickeners and emulsifiers in food products, as additives in mortar and concrete formulations or as a filler in pulp and paper manufacture. In binder formulations, it is usually employed as a thickening agent for other adhesives and has been known to be used as an ingredient in wallpaper pastes (Grossi, 1954) and as a digestible binder in pharmaceutical application (Guo et al., 1998).



Cellulose

Figure 1.14: Structure of cellulose (Pizzi and Mittal, 2003)

1.3.5.2 Chitin and chitosan

Chitins are polysaccharides that are extracted from the exoskeletons of crustaceans and insects. It can also be present in some forms of mould. Chitosan is created from chitin that has undergone alkali deacetylation. It is a heteropolymer composed of randomly distributed β-(1-4)-linked D-glucosamine and N-acetyl-D-glucosamine units (Figure 1.18). Chitosan films have been proven to be suitable bioadhesives for the use in pharmaceutical applications (Nunthanid et al., 2004), as well as for wound treatment and tissue repair (Dragostin et al., 2016; Mati-Baouche et al., 2014b). Chitosan has also found to be a suitable adhesive for metal. For example, high shear resistance of a chitosan adhesive with plasticizer was found when glued with chemically treated aluminium (Patel et al., 2013). For wood adhesion, a chitosan – laccase phenolic adhesive was formulated in the work of Peshkova and Li (2003), where the mechanism of adhesion was found to be similar to that of mussel proteins. Chitosan has also been used in the manufacture of biobased insulating composites, using sunflower stalks as reinforcement (Mati-Baouche et al., 2014a).

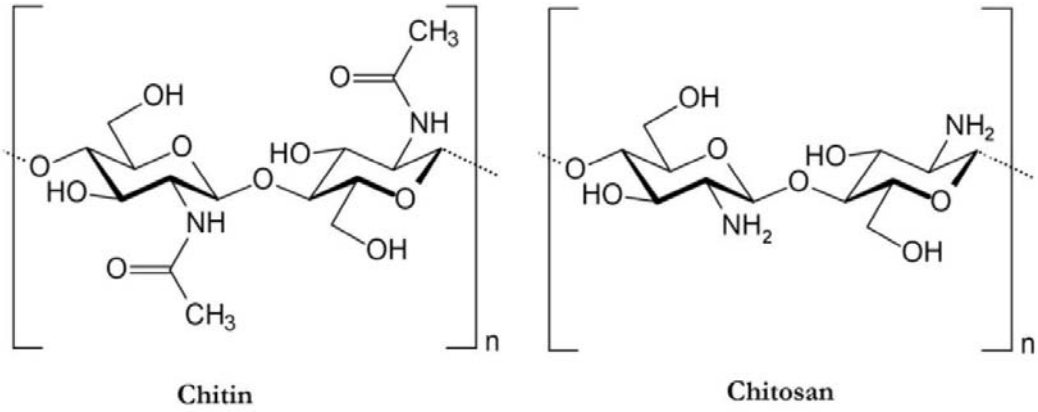


Figure 1.15: Structure of Chitin and Chitosan (Rinaudo, 2006)

1.3.5.3 Pectins:

Pectins are natural heteropolysaccharides. Their structure consists of linear 1,4-linked α-Dgalactosyluronic residues, obtained by extraction of certain plant material. It is a complex mixture of polysaccharides that makes up about one third of the primary cell wall of plants. An important organisational property of pectin is related to the esterification of galacturonic acid residues with methanol or acetic acid. This factor defines its gelling and adhesive performance. The degree of methylation is defined as the percentage of carbonyl groups esterified with methanol. If more than 50% of the carboxyl groups are methylated the pectins are classified as high-methoxylated pectins (HM), and less than that degree of methylation are classified low methoxylated (LM) pectins. HM-pectin requires a low pH of around 3.0, in order to form gels. HM-pectins are soluble in hot water and are prone to lumping without a dispersion agent. LM-pectins can gel at most pH levels, but require small levels of calcium for gelation (Sharma et al., 2006). Most commercial pectins are extracted from the peels of apples and citrus fruit, due to the fact that their molecular size and degree of esterification allow consistent gelling. Pectins have recently shown promise as a binder for pharmaceutical applications (Thakur et al., 1997). They are typically produced as water soluble brown, white, powders. Pectins are used as thickening and gelling agent in food products and is a main ingredient present in jams and jellies. Not much research has been made to integrate pectin as a binder in composites. In the work by Flory et al. (2013), commercial pectin was found to have the

same tensile strength as commercial synthetic binders for the application of air-laid pressed paper, which can be typically used for wet-wipe products.

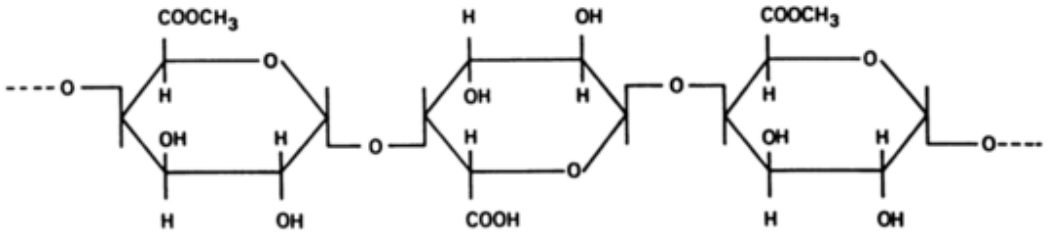


Figure 1.16: Partial structure of pectin (Baker et al., 2012).

1.3.5.4 Natural Gums

Gums are natural heterogeneous polysaccharides which increase the viscosity of a solution. Natural gums can either be produced by exudation, from either the stem or root of plants, from the endosperm portion of specific seeds. Gums have uses in a number of products: food thickeners, denture adhesives, pharmaceutical tablet binders, etc. (Conner, 1990). The most prevalent example is guar gum, coming from the endosperm of guar beans. They are galactomanns, which consist of (1-4)-linked D-mannopyranose chains linked to attached (16)-linked d-galactopyranosyl units (Figure 1.20). Guar gum has been known to be used in papermaking as a densifier and a method to improve stock retention (Larsson, 1988). It has also been known to be used as a tackifier for hydromulch erosion control to prevent clumping (Vaughn et al., 2013). Other examples include Arabic, locust bean, tamarind and Xanthan gums. Norström et al. (2014) studied dispersions of locust bean gum, guar gum, xanthan gum and tamarind gum as potential wood adhesives and found that locust bean gum had equivalent tensile strength to commercial polyvinyl acetate binder, despite low solids content due to high viscosities of the dispersions. Guar gum forms a thixotropic rheological system in a solution. Dilute solution at 1% concentration can have a high viscosity value of 10000 mPa.s (Mudgil et al., 2014).

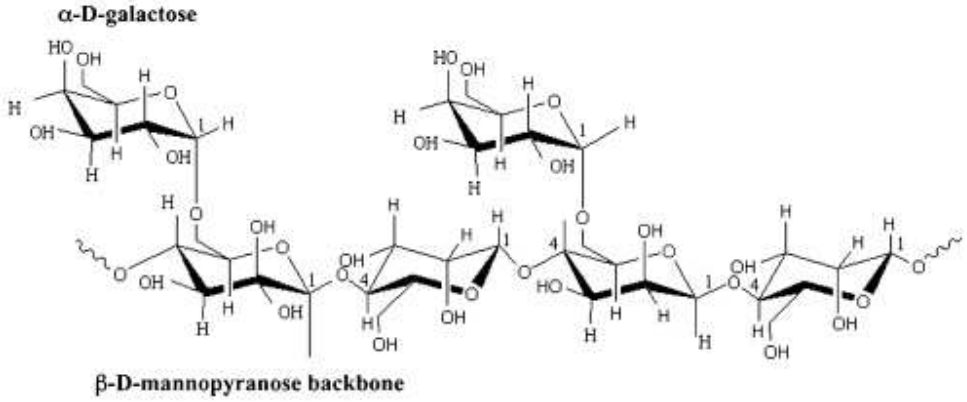


Figure 1.17: Structure of guar gum (Mudgil et al., 2014).

1.3.5.5 Starch

Starch is a carbohydrate polymer found in plants and crops such as corn, rice, tapioca and potatoes. The structure of starch is an arrangement of glucose units joined together by glycosidic bonds, forming two different molecules: amylose and amylopectin. The amylose molecule has a linear arrangement of glucose, while amylopectin has a more branched structure (Figure 1.17). Starch is typically composed 20 % to 30 % amylose and 70 % to 80% percent amylopectin. Starch very prominently used in food applications as a thickener and binder. Native starch is not soluble in cold water due mainly to its tightly-bound granular form. For native starch to be used as an adhesive, these granules must be opened up, which is attainable through numerous modification methods. These modification methods include: heating, alkali treatment, acid treatment, and oxidation. A formulation for a starch adhesive can include numerous additives to modify the adhesive's properties, such as borax to improve tack and viscosity, urea as a plasticiser, pol(vinyl alcohol) for water resistance, and bentonite as a filler, amongst others (Baumann and Conner, 1994). As an adhesive, starch is predominantly used in the paper industry. Starch can be mixed with cellulose pulp either in the wet end of paper manufacturing in order to improve dry strength of the paper (Floyd et al., 2001), or in the dry end to strengthen of modify the paper surface (Wagle and Yasnovsky, 1990). Starch adhesives are very thoroughly used for corrugated cardboard production (Czerwin, 1978; Fischer Jr and Mcelmury, 1969). For wood panel production, unmodified starch has

shown to have too low bonding properties and requires modifications of the polymer such as graft copolymerization (Wang et al., 2012; Wu et al., 2009; Zhang et al., 2015) or crosslinking (Imam et al., 2001, 1999). Some researchers have found modified starch to be a suitable binder for natural fibre insulating materials, such as insulation boards incorporating straw, corn pith and rice husks (Lacasta Palacio et al., 2014), and hemp fibre concrete, with starch as an alternative to the traditional lime binder (Benitha Sandrine et al., 2015; Le et al., 2014).

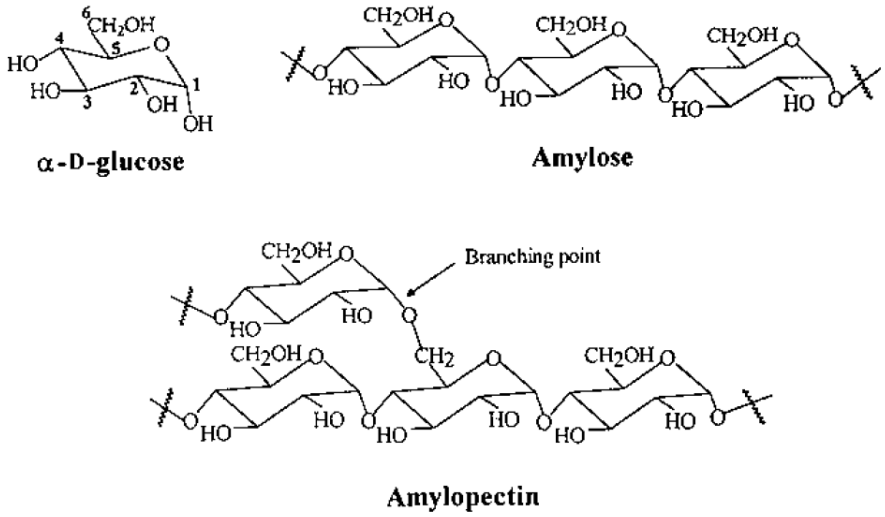


Figure 1.18: Structure of starch components (Pizzi and Mittal, 2003).

1.3.5.6 Lignin

Lignin is the second most prominent polymer found in trees, plants and other types of biomass. Within the cell walls of vascular plants, lignin contributes to plants' mechanical resistance and protection from its degradation. Most wood species contain 20% to 30% lignin. Its structure is composed of phenylpropane units joined with many different linkages. These units are shown in Figure 1.15.

During the manufacture of paper and paper products lignin is removed during the pulping and bleaching, making lignin by-products that could be used in adhesive applications. The main by-products are Kraft lignin (black liqueor) from the Kraft pulping process, and lignosulphonate from sulphite pulping. Lignosulphonate as an added binder has been known to be used in road formulations to reduce erosion. Since the

polyphenolic structure of lignin is similar to that of phenol–formaldehyde resins, lignin has shown the potential for use as a binder in the making of wood panels. For the manufacture of plywood, it has been found that the partial substitution of synthetic phenolic resins with lignin can produce plywood panels with satisfactory bond strength and water resistance properties (Klašnja and Kopitović, 1992; Mittal and Sharma, 1992). The same has applied for the manufacture for fibreboard and particleboard (Haars and Huttermann, 1984; Kazayawoko et al., 1992; Mancera et al., 2011), (Mancera et al., 2011). Despite these advantages, lignin-formaldehyde wood boards have not as of yet been considered commercially viable, mostly due to lengthy curing and pressing times (Pizzi and Mittal, 2003), therefore most of the spent lignin liquors in pulp mills are not recovered and burned as an energy source.

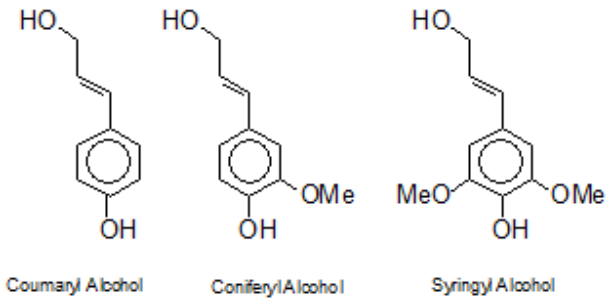


Figure 1.19: Structure of three phenyl propane monomers of lignin. (Hatakeyama and Hatakeyama, 2009)

1.3.5.7 Tannin:

Tannins are phenolic compounds that can be extracted from several types of wood, leaves and fruits. In trees and woody plants, tannins are present in the bark layer, protecting the plant from exterior attacks such as fungi and bacteria. Depending on the source plant, tannins can have varying molecular sizes and complexities. Most commercial tannins are extracted from the barks of Quebracho, Chestnut or Mimosa trees. Tannins come in two main forms: condensed and hydrolysable tannins. Most industrial tannins come from hydrolysable tannins due to their high reactivity. Its main use as an industrial product is mainly as a colorant and is utilized in the manufacture of tanning leather, and in ink formulations. As with lignin, tannin presents potential use as an adhesive due to its phenolic structure, which could be used to partially replace

synthetic phenol formaldehyde and urea formaldehyde binders. There exist numerous examples where tannins were used as in the formulation of binders for particleboard (Bisanda et al., 2003; Coppens et al., 1980; Kim et al., 2003; Pizzi, 1982; Pizzi and Merlin, 1981; Roffael et al., 2000). More recently, successful formulations for high density fibreboard incorporating cationic tannin were found to improve hardboard properties due to ionic interactions between the positively charged tannin and negatively charged wood fibres (Widsten and Kandelbauer, 2014). Hussein et al (2011), produced composite sheets with a Tannin-phenol Formaldehyde binder reinforced with cellulose paper pulp. Tannin and Tannin -Polyvinyl acetate blends have also shown promise as an adhesive in wood-based flooring (Kim, 2009).

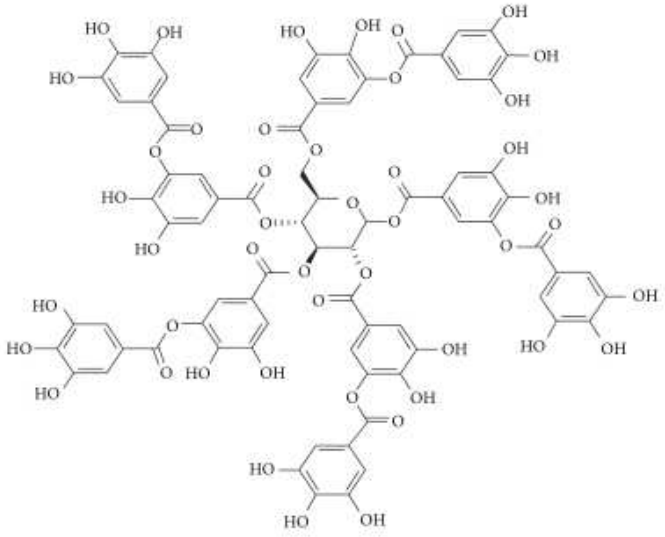


Figure 1.20**:** Structure of tannic acid. Tannin acid (Ahmad, 2014).

1.3.5.8 Protein glues

Proteins are linear polymers consisting of one or more long chains of L-α-amino acids, which are present in most living organisms. Protein adhesives can come from either plant origin such as soy proteins or gluten, or animal origin such as casein, gelatine or blood glues.

Casein is a protein adhesive made from milk. In order to make casein, defatted milk is precipitated in either acid at a pH of 4.5, or through enzymatic conversion of the milk's lactose to lactic acid. The precipitated curd is then washed to remove the acid and

impurities, dried and ground into a water soluble powder. In food formulations, casein is an additive in cheese and an important source of amino acids, carbohydrates, calcium and phosphorus. It has also been known to be used in paint formulations before acrylic paints became standard (Menaker, 1938; Scholz, 1953). In order to be made into a glue, casein must be mixed with an alkali, which is usually hydrated lime. Lime improves the adhesion, working life, and water resistance of the binder (Frihart, 2010). Other additives, such as sodium hydroxide are also included in the formulation to provide mould resistance. Casein is usually mixed with water and cured at room temperatures through solvent loss, though heat could be used in the curing and pressing process. Casein was once used for plywood manufacturing, but it became less favourable to soy and synthetic adhesives due to price and performance (Lambuth, 2003). Casein has however shown some niche applications such as for fire resistant panel and door manufacture (Selbo and others, 1960), and for bonding of wooden sports equipment (Bye, 1990).

Other animal glues originate from animal blood, bones or hide. Their adhesive properties vary depending on the animal source, such as cows, pigs sheep, fish etc. These types of animal glues are either derived from collagen, one of the major constituents of skins, cartilage and bones, or from albumin, a type of globular protein found in blood and egg whites.

Collagen involves a series of long protein molecules made up of naturally occurring amino acids which are linked by covalent peptide bonds. Fish collagen glues can be produced from the skin and bones of non-oily fish types and are sold in liquid form. As a commercial product, collagen glues are usually in a pure denatured form of gelatine. The denatured protein is spray dried in a fine powder. Upon dilution in water the powder forms a gel in ambient temperature, which when heated, turns into a tacky liquid which hardens as it dries. Animal bone and hide is mainly used in bookbinding, paper sizing, woodworking, and conservation (Pearson, 2003; Schellmann, 2007).

One benefit in using albumin blood adhesives is that they show higher water resistance than plant proteins (Raeker and Johnson, 1995). While blood adhesives could either be cold or hot pressed, bone and hide act as hot-melt adhesives. Recently, cow blood adhesive has been found to be suitable alternative for phenol formaldehyde through alkali modification (Lin and Gunasekaran, 2010).

Currently, for most uses, plant proteins are favoured over animal protein binders. Soy proteins were used as adhesives in the 1920s and 30s but fell out of favour after the development of petroleum based glues, which showed higher adhesive strength and better water resistance (Johnson et al., 1984), although recent advances in processing technology have created a resurgence in interest in soy protein as an adhesive. Soybean adhesives are produced from soybeans which have been dehulled, crushed and their oil is removed through high pressure or solvent extraction. This leaves a residual soybean meal which in turn, is used to create either soy flour (SF) soy protein isolates (SPI), or soy protein concentrates (SPC) (Kumar et al., 2002), which differ in protein, fat and oil content. As with other biopolymers mentioned beforehand, soy protein has shown to be a viable partial replacement with phenol formaldehyde, urea formaldehyde, and other synthetic resins for wood board manufacture, (Kuo et al., 1998; Lorenz et al., 1999). Other soy-protein blends that have been studied include soybean-blood (Brother and Smith, 1940) and soybean-casein glues (Bradshaw, 1931). While these blends have shown positive features in wood manufacture, price and long pressing times make them impractical. In order to have 100% soy protein adhesive formulations, the adhesion and water resistance properties of the protein need to be improved through chemical or enzymatic modification. Current modification strategies include protein denaturation, crosslinking, enzyme modification, and chemical reactions such as acylation and oxidation. The works which have applied these modification methods and their results were reviewed by (2002). Aside from wood binding, Soy protein adhesives have also been known to be used in the manufacture of cardboard (Zhong et al., 2001), paper coating (Krinski et al., 1990, 1987), and low density particleboard from agricultural residual fibres (Wang and Sun, 2002).

Another plant protein that has good binding potential is wheat gluten, an industrial by-product from the processing of wheat starch. Modified wheat gluten has recently also shown promise as an adhesive in the manufacture of plywood and particleboard (Khosravi et al., 2015, 2011; Vo Hong et al., 2015)

1.3.6 Surfactants

Surfactants are compounds that can modify the properties of a liquid/liquid, solid/liquid and gas/liquid interface. They are usually used to change the surface tension (or interfacial tension) between two liquids or between a liquid and a solid. They are used in many applications as detergents, wetting agents, emulsifiers, foaming agents, and dispersants. They are amphoteric which means that they contain both hydrophobic groups and hydrophilic groups, also known as the "head" and the "tail" respectively. These groups make them soluble in both organic solvents and water. When in contact with water, surfactants form aggregates, with an inner core formed by hydrophobic tails of the surfactant and the groups form an outer shell from the hydrophilic head. These aggregates are known as micelles, for these micelles to be formed, a minimum surfactant concentration, known as the critical micelle concentration (CMC), must be reached.

For cellulose insulation, surfactants can contribute to the decrease in drying times through two mechanisms. Firstly the reduction of water/air surface tension and cellulose/water contact angle of the sprayed water can enhance water removal from capillaries within fibre walls and between fibres. Secondly the adsorption of cationic substances on negatively charged cellulose has been well documented (Alila et al., 2005). Cationic surfactants have found to modify the surface properties of cellulosic fibres and directly influence the papermaking and deinking processes on a large scale of paper pulp (Beaupré, 2012), and a similar outcome could be reached with cellulose insulation, to modify its interactions with water.

1.3.6.1 Types of surfactants

. One can catalogue surfactants according to the charge of their hydrophilic portion as: anionic, cationic, amphoteric (or 'zwitterionic') or non-ionic surfactants.

Anionic Surfactants: contain a negatively charged hydrophilic head, usually combined with an alkaline metal such as Na+, K+ or a quaternary ammonium. Their main uses are in detergent formulations due to the fact that the negatively charged head group can be repelled from most negatively charged surfaces, and thus lift particulates. They are the most inexpensive, commonly used surfactants and represent about 50 % of the market

for surfactants. Examples include alkyl sulphates, sulphonates, alkyl carboxylates and fatty acids salts.

Cationic surfactants: have a positively charged hydrophilic head. Due to their affinity towards negatively charged surfaces such as hairs, fabrics and bacterial membranes, their positive charge allows them to be used in fabric softeners, hair conditioners, corrosion inhibitors and antibacterial agents. They are less prevalent due to their high cost of the high pressure hydrogenation during synthesis. Fatty amine salts and quaternary ammonium salts are examples of cationic surfactants.

Non-ionic surfactants are surfactants whose hydrophilic head has a neutral charge. They can usually be found in formulations of emulsifiers, dispersants and low temperature detergents. They are the second most prevalent kind of surfactant. Some non-ionic surfactant include sucrose esters, polyol esters and polyoxyethylene esters

Amphoteric or Zwitterionic surfactants exhibit both anionic and cationic head groups, with charge highly dependent of the pH of the solution. They can be used in shampoo and cosmetic products due to their high foaming properties and their low irritation qualities. Examples include betaines or sulfobetaines and certain amino acids.

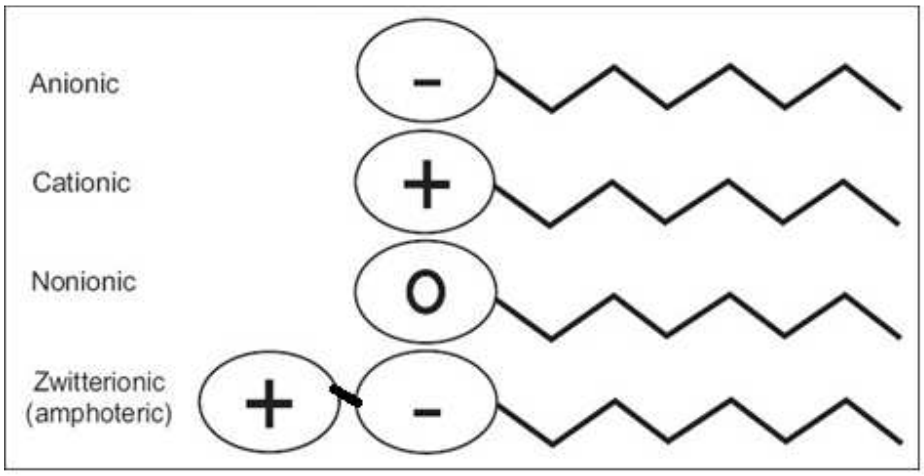


Figure 1.21: Schematic representation of types of surfactants with hydrophilic head and hydrophobic tail.

1.3.6.2 Surfactants as dewatering agents in cellulose

The work of Beaupré (2012) Studied interactions of cationic surfactants during the papermaking process. His work found that the addition of a cationic surfactant cetyltrimethylammonium bromide (CTAB) to a paper pulp slurry contributed to the

increase in the removal of water during the paper forming and pressing sections, thus requiring less energy and time for drying. It was found that the addition of CTAB reduced the water retention and modified the pore structure of the negatively charged fibres. Some detrimental effects of the addition of CTAB were also encountered: the mechanical strength of the paper sheets was reduced and the surface charge of the fibres was reversed from negative to positive. Many different varieties of cellulosic pulp were tested. Figure 1.22 shows a 15% decrease in water retention value (WRV) of recycled newsprint paper pulp, the same source material as cellulose fibre insulation. While still taking into consideration differences in water dosage and processing conditions, this decrease in water retention could be extrapolated to a reduction of CFI drying times.

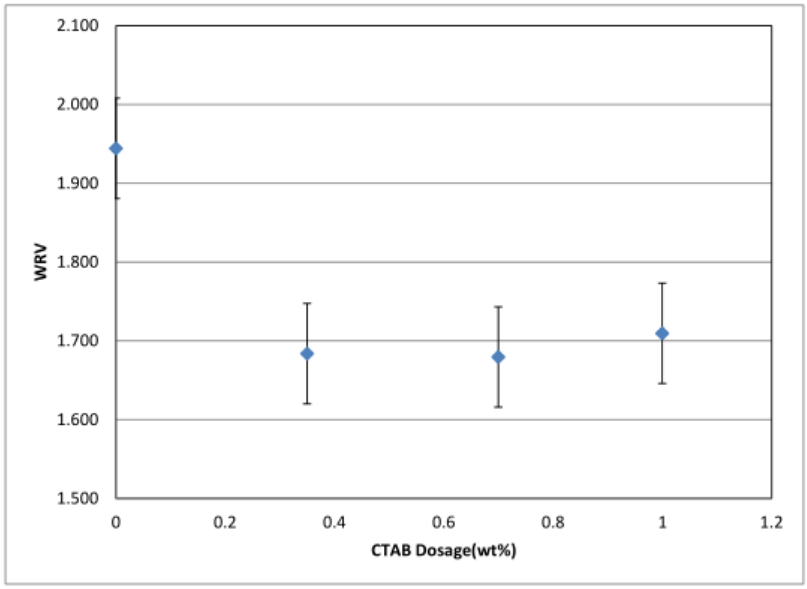


Figure 1.22: Water retention value vs CTAB dosage in recycled newsprint paper pulp (Beaupré (2012)

1.4 CONCLUSIONS

As has been shown by the available literature, the building sector has a strong effect on worldwide CO_2 emissions and energy consumption. Two methods to reduce the environmental impact of the building sector include, the increase in the insulation requirement of new and existing materials, and the use of alternative building materials comprised of natural or recycled materials with low embodied energy and ecological impact. CFI is an innovative eco-friendly insulation material that presents similar

characteristics in terms of thermal comfort and performance to its non-renewable counterparts. Nevertheless the material presents, in its current state, some disadvantages compared to less eco-friendly insulation materials and has shown the need for more optimization and development. Further research needs to focus on studying and resolving the issues with the material's properties and performance.

Although the current available research thoroughly presents the properties of CFI as installed through the dry methods (which have historically been the most prevalent installation techniques), there is little current information on the wet spray method. One work has shown the main issue with the wet spray method: the weather-dependent drying times of the material. The study also demonstrated the variability in the installed moisture content, which also leads to increased drying times. No research has yet determined the effect of the applied water on the thermal conductivity of cellulose insulation, or characterized the material's mechanical performance when applied with water. The next chapter will confront this issue.

The current research on adhesives composed of natural polymers shows the variety of potential candidates that could be used as a biobased additive with wet sprayed cellulose insulation. While there are numerous examples of biobased adhesives being used in the manufacture of natural fibre composites such wood fibreboards, paper pulp, and hemp concrete, the manufacture of these composites differs somewhat from that of wet spray CFI. Most notably, the specific conditions in which wet-spray cellulose fibre insulation is manufactured and applied will define the nature of the biobased additive to be used. Most of the cited examples have specific temperature, pressure, and/or material requirements that are not compatible with the wet spray method. As an example the use of a thermosetting polymer would be unfeasible since it would be extremely difficult to heat a polymer during the spraying process, not to mention the increased energy costs, so is the suitable candidate adhesive should be a solvent loss adhesive that hardens with the removal of moisture at ambient temperature.

As has been shown, the mechanics which define the process adhesion are complex and varied. Taking into account the adherent (loose cotton like porous cellulose fibres), the optimal binder will react and adhere with cellulose fibres through mainly

adsorption (hydrogen bonds), with the mechanical and diffusion mechanisms also taking place. These optimal properties are detailed more in depth in chapter 3.

With regards to the reduction in the drying times, four scenarios could be envisioned through the use of biobased additives:

- The moisture sorption and subsequent curing of added adhesive accelerates the evaporation rate of water, thus making the sprayed CFI dry faster.
- The increased mechanical resistance of CFI due to the addition of a binder decreases the required initial water dosage, thus making the material reach a dry state sooner.
- The increased solids content of the CFI with the added additive decreases the moisture to solids ratio, therefore making the material initially drier then with just water.
- The decreased surface tension of the water through the use of a surfactant allows faster flow of the water through the pores of the material, thus decreasing drying rates. The absorption of cationic surfactants through the negatively charged fibres could also have an influence.

These additives need to improve these properties without being detrimental to the performance of cellulose fibre insulation. Mainly the density and thermal conductivity of the material need to remain low, but also the fire and fungal resistance of the material must not be affected. It is through these innovations that cellulose fibre- based insulations can become more prevalent and contribute to more eco-friendly construction projects.

2

THE PROPERTIES OF CELLULOSE INSULATION AND THE THEIR RELATION TO WATER DOSAGE

2.1 INTRODUCTION

The available information of the state of the art has shown the benefits and downsides in the use of cellulose insulation for new constructions and refurbishments. However, research has mostly focused on the properties of dry blown cellulose. Little work has been done to characterize the raw material (loose recycled newsprint fibres), before installation. While some parallels could be made with the manufacture of recycled newsprint paper pulp, the method of manufacture and final product of both materials vary greatly. In this chapter we aim to study the physical and chemical properties of recycled fiberized newsprint fibres, in order to better understand their behaviour as a final product: a rigid, self supporting insulating material. In order to do this, the shape and structure of fibres were first observed using microscopic and particle size distribution analysis. The degree of refinement of the fibres was studied using the Schopper-Reigler method. Chemical analysis consisted in the determination of lignocellulosic components within the fibres via the Acid detergent fibre (ADF) and neutral detergent fibre (NDF) method. Finally the apparent density of the material was determined via the tapped density method.

In order for a new formulation of wet spray CFI to be conceived, the traditional method of application needs to be studied. This includes not only the behaviour of the newsprint fibres with water, but also the influence that varying water dosage could have on the final properties of the insulation material. The interactions of the fibres with water were first considered with dynamic vapour sorption (DVS), water retention value (WRV) measurements, and differential scanning calorimetry (DSC) testing to determine bound and unbound water content. The drying behaviour of projected cellulose insulation samples was then investigated. The influence of water dosage on the thermal and mechanical properties of cellulose fibre insulation manufactured through the wet-spray method was studied. A method to test the compression resistance of the cellulose fibres was devised to study the influence of initial moisture content on its mechanical properties. The same was done with thermal conductivity testing via the guarded hot plate method. The results from these characterization tests provide a solid foundation in order to improve upon the properties of CFI via a new formulation.

2.2 PROPERTIES OF CELLULOSE INSULATION FIBRES

2.2.1 Origin and preparation of raw materials

Treated and untreated cellulose insulation samples (Figure 2.1) of the brand Univercell Comfort were provided by SOPREMA (Bordeaux, France). The raw materials are pre-consumer newsprint paper from several providers from the south of France. In factory, the papers are ground into smaller fragments, which are then turned into loose fibres via a specialized fiberizer. Boric acid and magnesium sulphate then added as a fire retardant and antifungal agent at proportions of around 15% the mass of CFI. The fibres come compacted in 12 kg bags in order to optimize transportation costs. For characterization of loose cellulose fibres, samples were taken at random points of a bag and separated into loose cotton-like flakes either by hand or using compressed air to separate the fibres. The loose fibres were then stored in a humidity chamber at 60% Relative humidity and 25°C. Moisture content of samples stored in these conditions as determined by EM01, ranged from 8% to 11%. In order to have a point of comparison for

some tests, treated cellulose insulation of the brand Ouateco (Saint Geours de Maremne, France) was also acquired.



Figure 2.1: Loose Univercell CFI fibres

2.2.2 Microscopic observation

Figures 2.2 and 2.3 show loose CFI fibres under different levels of magnification, as was done with experimental method EM02. As one can see the fibres vary in size, ranging from the micrometer scale to fibres less than 5mm in size. The inks in the fibres are visible, as are the added powdered additives on the surface. The fibres show a high presence of hairs and fibrils. This is known as fibrillation and is the result of the shearing and compressive forces during shredding via a specialized high speed fiberizer. This is what gives the fibres their lightweight, fluffy properties. If fibres were to be shredded via traditional shredding methods used in papermaking such as through a hammermill, they would have less of these microfibrils, thus making the material denser and thus increasing its thermal conductivity. When the fibres are applied via the dry method (blown in cellulose), the fibres expand with the applied air pressure but then consolidate when applied within a confined space (usually an attic or a closed wall cavity). Upon consolidation the fibres and fibrils interlock with each other, forming a self sustaining structure. In the wet spray method, the use of water reinforces this structure, making the final product more resistant to settling.

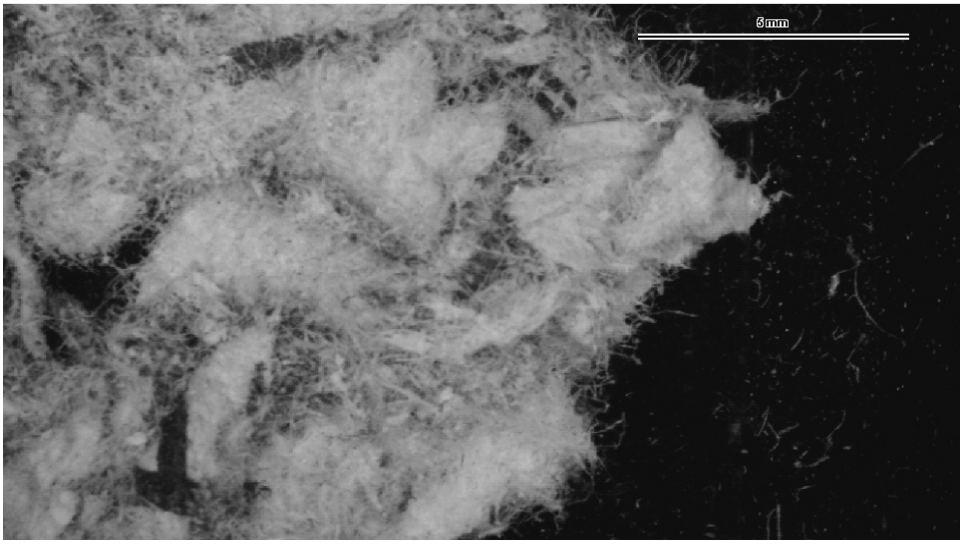


Figure 2.2 : CFI fibres under X7.5 magnification

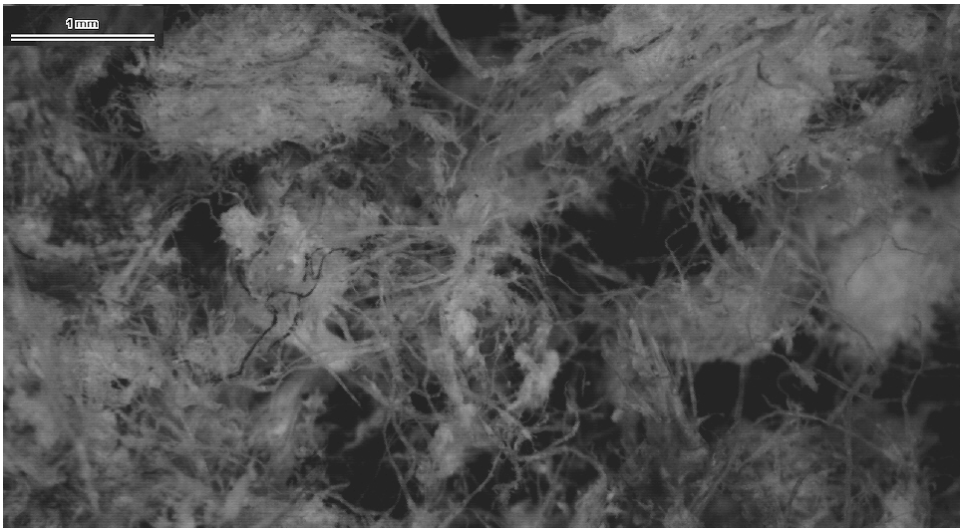


Figure 2.3: CFI fibres under X20 magnification

2.2.3 Schopper Reigler freeness testing

The Schopper Reigler test, as detailed in EM03 is a method to characterize and surface properties of cellulosic fibres. It is typically used to determine the rate of drainage of a pulp suspension during the beating and refining process of paper pulp manufacture. It is measured in a 0°SR to 100 °SR scale. It is determined by reading the inverse of the amount of the liquid collected as the pulp flows through the apparatus, divided by 10.

$$SR=V_{pulp}/10$$

(2.1)

The higher SR value, the more the pulp can retain water and the higher it has been refined. For cellulose insulation it is helpful in characterizing the swelling of the fibres in the presence of water. Samples of both brands of treated cellulose insulation fibres were disintegrated and turned into homogeneous slurry at 0.2% consistency. Table 2.1 shows the Schopper Reigler freeness for both brands of CFI with. No difference was found between untreated and treated cellulose fibres. Three pulp suspensions for each brand were measured and averaged, with standard deviation shown in parenthesis.

Fibres	Temp °C	Water collected in drainage cylinder (ml)	Schopper Reigler freeness (° SR)
Univercell (untreated)	21	580	42 (±3)
Ouateco	20	640	36 (±2)

Table 2.1: Schopper-Reigler freeness testing

The standard ISO 5267-1 requires the pulp suspension to maintain a temperature of 20°C which, for Univercell fibres was slightly higher. While this could have an effect on results, it has been shown that for a similar type of test, the Canadian standard freeness (CSF) a variation of ±5°C represents a variation of 10% of measured freeness (Gharehkhani et al., 2015). Both values of freeness were found to be close to 40°SR which is the value given in literature for recycled paper pulp. Standard deviation for both types of fibre was under 4, a requirement for results to be valid according to the standard. It is difficult to determine how this freeness value can affect the performance of wet sprayed CFI, especially since a collection of slightly moistened fibres is much different than a pulp suspension. Nevertheless these results give the indication that the method of manufacture of the cellulose fibres gives them similar swelling characteristics than those of recycled paper pulp.

2.2.4 Tapped density and loose fill settling

The cellulose fibres manufactured for the use in CFI have a high compressibility which makes them susceptible to settling, as has been shown by Svennerstedt (Svennerstedt, 1995). For the loose fill application the air pressure applied compacts the material to a sufficient density to prevent settling and ensure the material fills up the

volume either in a wall cavity or an attic without leaving spaces in the envelope in which cold air could infiltrate. For the wet spray application, the pulverised water provides an additional pressure which further compresses the material.

This compressibility is due to air voids, either between the cellulose fibres or within the pores of the cellulose. It is therefore important to distinguish between the bulk density and the compacted density of the material. The standards for loose fill cellulose insulation require settling tests to determine the compressibility of the material. These tests subject a sample of loose fill cellulose to cyclical impact tests to represent the settling of the material over time. The material for these tests is quite specific, so an alternative test was designed using a tapped density apparatus for powders, as described in EM04. Results for density before and after compaction of 1250 taps of treated and untreated Univercell and treated Ouateco samples, as well as their compressibility ratio, are shown in Table 2.2.

	Initial density (kg/m^3)	Tapped density (kg/m^3)	Compressibility (%)
Univercell (untreated)	21.3	43.1	49
Univercell (treated)	24.7	45.3	46
Ouateco	17.3	47.9	64

Table 2.2: Tapped density measurements.

The initial density presented through these tests are very low when compared to those made to traditional settling tests in the literature (30-40 kg/m^3) This is due to the fact that the loose fibres adhered to the cylinder of the tapped density apparatus. Once compacted, the densities more closely resembled those found through the traditional method. As a consequence the compressibility ratios are lower than through traditional methods. This makes the tapped density technique a viable method for determining settling if the initial loose density is already known, by weighing a sample of loose CFI fibres in a recipient with a known volume. Untreated samples had lower densities than treated samples which would be logical since the included additives weigh down the material. The higher compressibility between samples could make the Ouateco brand cellulose have a higher final density after spraying.

2.2.5 Particle size distribution

The particle size distribution test is used to determine the different mass proportions of particles of varying sizes of a certain material. It is measured in the cumulative mass of the particles of the material that pass through a series of metal sieves with successively decreasing hole diameter. Particle size distribution of both Univercell and Ouateco samples were determined according to the standard defined in experimental method EM05. The difficulty associated with this test was the separation of the cottonous flakes into individual fibres and fines. Compressed air was blown into the fibres to make them separate and pass through the sieves. Results for both brands and untreated Univercell are shown in Figure 2.4. One remarkable result is the high presence of fines, especially for the Univercell brand samples. In papermaking, fines are particle fragments of fibre walls with size less than 0.3 mm. These fines could also be the powdered antifungal and fire resistance additive that separated from the fines, as can be seen with the difference in amount of fines between treated and untreated samples. Usually recycled paper contains less fines than paper from virgin pulp (Wistara and Young, 1999). These fines are a result of the shredding of recycled newsprint during cellulose insulation manufacture. During the fiberization phase (the second size reduction) these fines are collected and reintegrated with the fibres, at the same time as boric acid and the additives are added. These fines could have an effect on the drying and mechanical properties of cellulose, but also increases its density, making the material more expensive to install and potentially increasing its thermal conductivity.

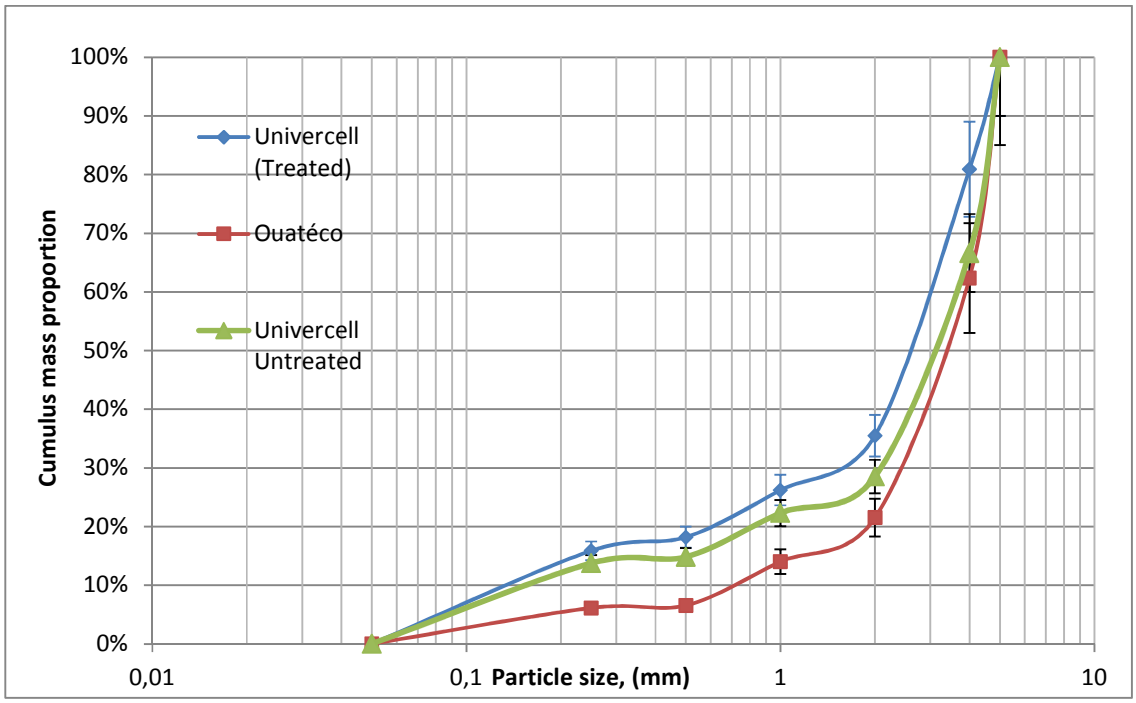


Figure 2.4: Particle size distribution of cellulose samples.

2.2.6 Chemical composition analysis:

As has been stated in chapter 1, the main raw material of cellulose fibre insulation is recycled newsprint with added fire and fungal resistant additives. Newsprint is composed mainly of mechanical pulp, and additives such as ink, kaolins and proteins. Mechanical paper pulp contains cellulose, hemicelluloses and lignin, with lignin content being higher for mechanical pulp than for chemical pulp. Cellulose and hemicellulose are hydrophilic and lignin is hydrophobic, so the proportions of these constituents within the fibres can potentially dictate their behaviour with water. The method to determine these proportions was defined by Van Soest and Wine (1968) and is detailed in experimental procedure EM06. The proportions of cellulose, hemicelluloses, lignin, ashes, and other constituents of Univercell and Ouateco brands of cellulose insulation are presented in Table 2.3. All percentages are expressed in relation to dry mass of the material, with moisture content of all samples ranging from 9% to 11%. The proportions for untreated samples are somewhat similar to those presented for newsprint in the work of Wu et al. (2001). The differences between the “others” content (mass of materials removed during neutral detergent treatment), could mean that the soluble fire and fungal treatment

additives are removed during this treatment. The differences in proportions of additives, and hydrophilic or hydrophobic constituents of both brands could influence their drying behaviour. The higher cellulose contents with Ouateco compared to Univercell brands could make higher moisture sorption capacity, thus slower drying times. It is however, important to note that the source material is inconstant. Different cellulose insulation manufacturers have different paper providers, all with different mechanical pulp formulations. These formulations could contain different proportions of recycled pulp, chemical pulp, different additives and/or different methods of manufacture. In the future these factors could change, making it quite difficult to optimize the raw material (recycled newsprint paper) in order to improve drying.

	Univercell (treated)	Univercell (untreated)	Ouateco (treated)
Cellulose	34.7%	39.9%	45.3%
Hemicelluloses	16.8%	17.3%	14.3%
Lignin	20.4%	29.8%	25.3%
Ash	3.5%	5.4%	1.0%
Others	24.6%	7.7%	14.0%

Table 2.3: Chemical composition of cellulose insulation samples

2.2.7 Interactions between water and cellulose insulation

2.2.7.1 Water retention value

The water retention value (WRV) of fibres is the measurement of the capacity for paper to hold water. It is measured through the removal of water via centrifugation. Around 5g of treated and untreated Univercell were analysed according to the procedure, adapted from standard ISO 23714 is described in EM08. The term “hornification” is used to describe a reduction in the amount of water that can be held within the fibre walls due to a transformation of the paper pulp. This mainly refers to fibres that have been refined once again or recycled, and for which external fibrillation has been exterminated, but it has been shown that drying of fibres can induce hornification as well (Hubbe et al., 2007). WRV testing could be a good technique of characterizing the changes in the properties of CFI during drying, since hornification through drying is thought to be an effect of pore closure and the strengthening of fibres. This theory is heavily debated, with

other theories including the formation of irreversible hydrogen bonds and the formation of covalent bonds within carbohydrate chains from within the fibres (Brancato, 2008). In order to attempt to characterize the hornification of wet spray cellulose fibre insulation after drying, some samples were first sprayed with pulverized water, of around 80% of their dry weight, including the ambient moisture already in the fibres. This could be considered an exaggerated amount of water dosage for the wet spray method, but it is necessary for the amount of hornification during drying to be characterized. The average in WRV of 3 samples for each type are shown in Table 2.4

Sample	WRV g/g	Standard deviation coefficient
Univercell (treated)	2.25	15%
Univercell (untreated)	2.16	9%
Ouateco (treated)	2.34	11%
Univercell (treated, air dried from 80%)	2.04	13%

Table.2.4: Water retention value (WRV) of cellulose insulation samples

The values of WRV found are much higher than those cited in the ISO 23714 standard for recycled paper pulp (1.6 g/g), but closer than those cited by Beaupré ((2012)) (see Figure 1.21 in Chapter 1) for recycled newsprint. Generally, the higher the WRV the more energy will be required to dry the paper. It is also an indication of the high content of mechanical pulp, where the values of WRV reach around 1.9 vs chemical pulp which are around 1.2, depending on refining conditions. This could be due to the fact that the filtration method used is different than that specified in ISO 23714, making some of the water hard to remove from the fibres during centrifugation. Treated cellulose fibres seem to have a higher WRV than untreated fibres, which would be logical since the included additives are hygroscopic and would absorb some of the water. Redried fibres showed some a slight reduction in water retention, although the high variability of samples makes it hard to state a conclusion. While high, this variability is typical for WRV of recycled pulps. In fact, it has been shown that mechanical and thermomechanical pulps are highly resistant to hornification. This could be due to the presence of lignin in the structure, which interfere between interfibre hydrogen bonding (Diniz et al., 2004). The slight

difference between WRV of both brands of CFI could be an indicator of drying behaviour.

2.2.7.2 Bound water determination with differential scanning calorimetry (DSC)

The water within cellulose fibre walls can be catalogued in three different categories. Non-freezing bound water is the water present in the micropores of the fibre. It corresponds to the water bound to the hydroxyl and carboxylic groups in pores of the fibre. The freezing bound water appears in pores larger than micropores and binds to hemicelluloses. As the freezing bound water dries, this causes the pores of the fibres to close irreversibly. The free or unbound water is the water present in the larger pores and between the lumen in fibres. Figure 2.5 shows the interactions between these types of water and cellulosic fibres.

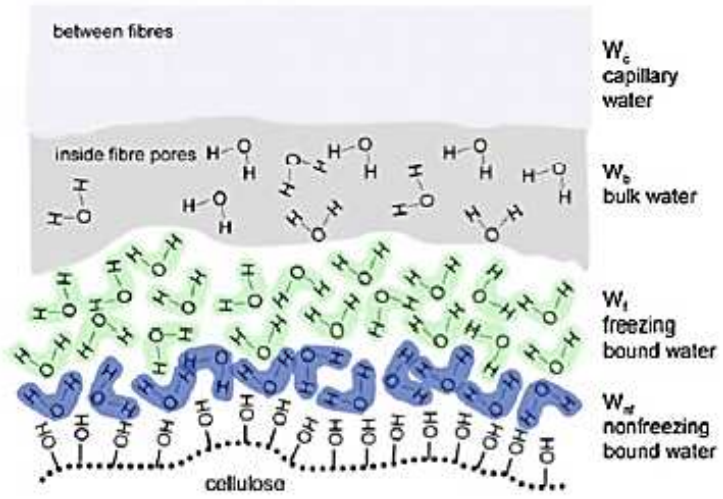


Figure 2.5: Types of water present in cellulosic fibre surface (Samyn, 2013).

In order to characterize these kinds of water, within the fibres a method was developed by Nakamura et al. (1981) . The full method is described in EM08, and the results are shown in Figure 2.6.

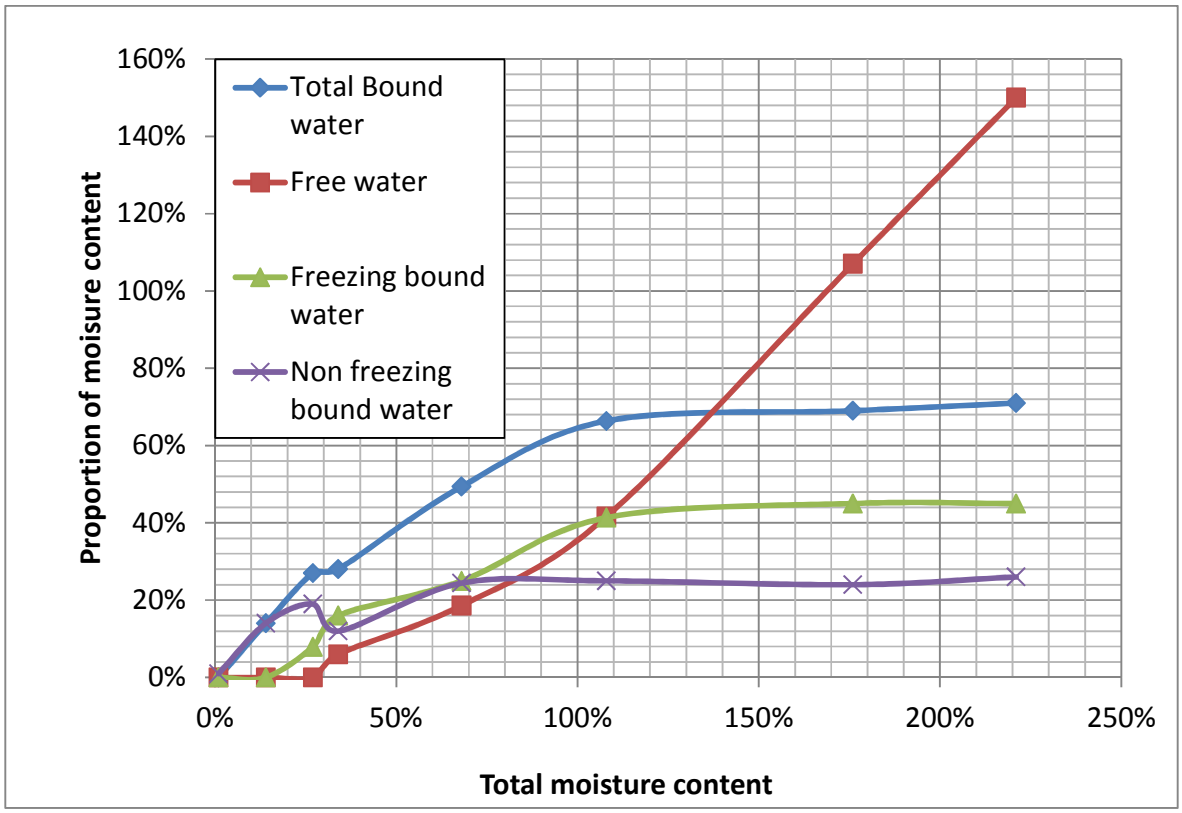


Figure 2.6: Frozen and unfrozen bound water, free water and total bound water as a function of total water content for untreated Univercell CFI.

The Figure shows how for low moisture contents, only non-freezing bound water is present, then as moisture content increases, other types of water start to appear: at around 14% for freezing bound water and 27% for free water. The values of both freezing and non-freezing bound water increase gradually until the micropores are saturated. It is at this point, at around 108% moisture content, in which free water fills the bigger pores and the voids between fibres. Past this point only the amount of free water increases. For the recommended dosages of cellulose insulation of 40 to 60% moisture content, mostly bound water is present, with smaller proportions of free water, which increases as total moisture content increases. Once the fibres are sprayed, the bound water strengthens and closes the pores within the fibres while the free water reinforces the link between fibres and microfibrils. This only applies once the water is fully absorbed within the fibres, as during spraying the bound water in the surface of the fibres gradually penetrate into smaller pores. The presence of freezing bound water in this dosage range means that as the fibres dry, the size of pores diminishes and this change in pore size could affect the

thermal conductivity of the material. As the fibres dry, the free water is the first to diffuse within the material and evaporate since it is more present in the surface of fibres. For the equilibrium moisture content of 8% to 11% at 60% humidity after drying, only non-freezing bound water is found in the material. As was shown by Weise et al. (1996), one can correlate free water from hornification values, so the amount of free water increases, so does the WRV.

2.2.7.3 Dynamic vapour sorption (DVS)

DVS testing was done according to EM07. Results from dynamic vapour sorption of cellulose fibres are shown in Figure 2.7. The isotherms show little hysteresis, less than 2% difference between sorption and desorption isotherms. In the hygroscopic range moisture sorption reaches a maximum of around 20%. This is close to the result for DSC for when free water starts to appear (27% moisture) .

It is possible however, to estimate the moisture sorption in the whole range using mathematical approximations. Many models exist such as Brunauer–Emmett–Teller (BET) or Guggenhein, Anderson, de-Boer (GAB) models, but for porous solids such as cellulose it is recommended to use the Oswin model (Barreira et al., 2014):

$$X_e = b_0 \exp\left(\frac{b_1}{T}\right)\left(\frac{a_w}{1-a_w}\right)^{b_2}$$

(2.1)

Where X_e is the calculated moisture content, a_w the relative humidity, T is the ambient temperature, and b_0, b_1, b_2 are coefficients dependent on the material. These coefficients were determined by minimizing the standard deviation between the calculated values and the average between the measured sorption and desorption values. The Oswin model shows good relation with the values determined experimentally. The moisture content at saturation can be estimated when RH values approach 100%. At a RH trending towards 100% the Oswin model gives a value of 279% moisture content, showing that, past the hygroscopic range, cellulose can absorb high quantities of water via capillary conduction. For cellulose insulation drying, this would mean that the range of minimum applied water dosage of 40%-60% corresponds to water past the hygroscopic range and within the capillaries. In practice, capillary water tends to dry

faster than absorbed water within the pores although below the threshold of 20-15% moisture content, the cellulose insulation is already considered dry. As was presented in chapter one, this high hygroscopisity is also considered favourable for building envelope materials, as it makes the material "breathable" and thus lower ambient humidity in a room to a comfortable level, or conversely increase it when it is too dry.

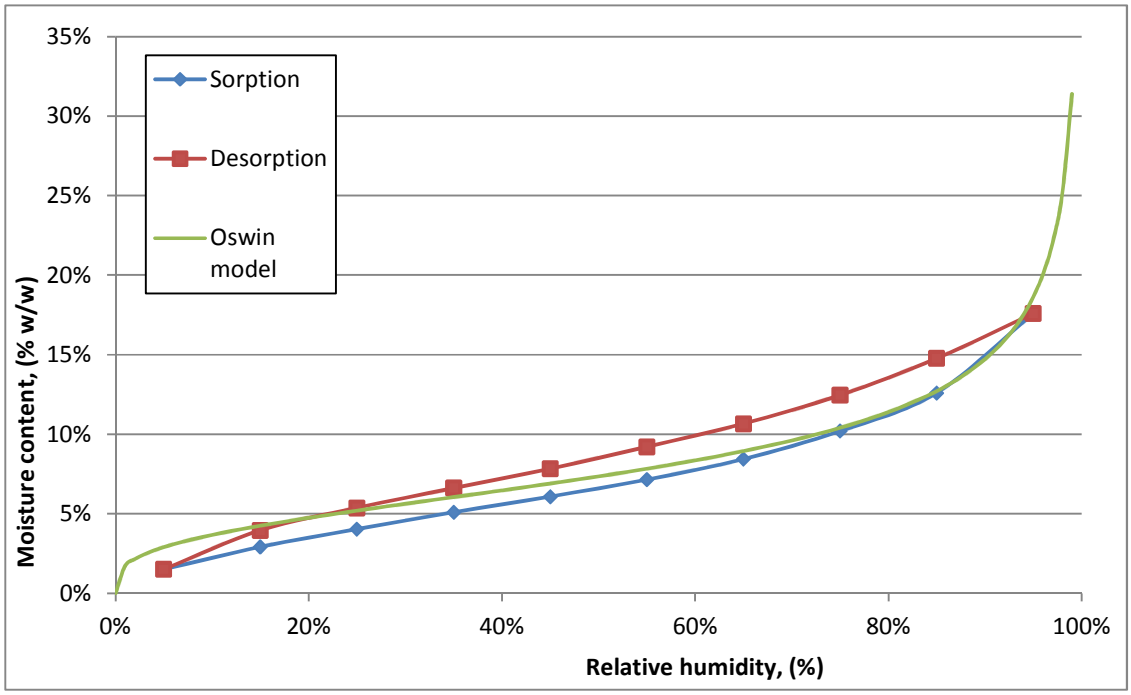


Figure 2.7: Dynamic vapour sorption of CFI, with Oswin model approximation.

2.2.8 Drying of cellulose samples

Wet spray cellulose samples were sprayed and stored according to the method described in EM10. To measure the drying of CFI, the mechanical specimens were weighed daily in order to measure the evaporated water until equilibrium moisture conditions at 60% RH (a mass variation of less than 1% in 24 hours) were reached. The equilibrium moisture content was then determined by taking 3g of fibres from the ambient dried sample and drying them at 103°C for two hours, as described in EM01. Using the mass of the samples and the equilibrium moisture content, the initial moisture content as well as the drying of the material at 24 hour intervals was determined. Moulds with the minimal moisture content of 40% were also sprayed at higher thicknesses and

compacted after drying to give denser materials with the same initial moisture as a reference point (see EM10).



Figure 2.8: Projected CFI moulds.

2.2.8.1 Influence of treatment and source material

An objective of this work is to investigate cellulose insulation with different properties and how those properties could influence its drying behaviour. The physical and chemical differences between brands of CFI fibres, as well as their potential influence on drying are summarized in Table 2.5. Given the measured differences in properties, it would be probable that Univercell treated brands might be able to dry faster.

Property	Brand		Potential influence on drying
	Univercell	Ouateco	
Particle size distribution	More fines	Less fines	Fines absorption could make the material dry faster
chemical composition (ADF/NDF)	Less cellulose and hemicelluloses	More cellulose and hemicelluloses	Higher hygroscopic cellulose content could make the material dry slower
Tapped density	more compressible	less compressible	More compressible materials could potentially dry faster, due to ease of moisture transport.
WRV	Slightly lower	Slightly higher	Higher WRV suggests slower drying.
Schopper Reigler	Similar values	Similar values	Higher SR could mean faster drying, as with paper production.

Table 2.5: Measured differences between cellulose brands and possible influences on drying.

In order to confirm this hypothesis, untreated and treated Univercell, and treated Ouateco samples were sprayed on the standard 300x300x90mm^3 moulds at around 65% moisture content, and the evolution of this moisture content was studied. As can be seen in Figure 2.9, Ouateco samples dried slightly slower, as was predicted. At 72 hours, a difference of 8% moisture content could be measured between both brands. Untreated fibres dried slightly slower than treated fibres, but equilibrium moisture content was lower. The differences in drying between untreated and treated samples are clear; the additives help absorb the moisture and are within the water, but as the material dries, the additives retain some of the sprayed moisture.

One would assume that the differences in the measured properties of the fibres are the cause for the differences in drying. There are two major limitations to this line of thinking: one, as mentioned previously, the sources of newsprint have variable methods and source material for manufacturing its paper, so it would be hard to ensure that the material continuously conserves its physical and chemical properties. The other is the fact that the specific proportions and composition of the added antifungal and fire resistant additives are unknown and confidential. As we were unable to obtain untreated CFI samples of the Ouateco brand, the differences between untreated samples could not be

asserted. The contribution of these hygroscopic mineral additives to moisture absorption and subsequent drying could take priority over the influence on the studied properties of the fibres. Nevertheless these are factors to consider in terms of quality control.

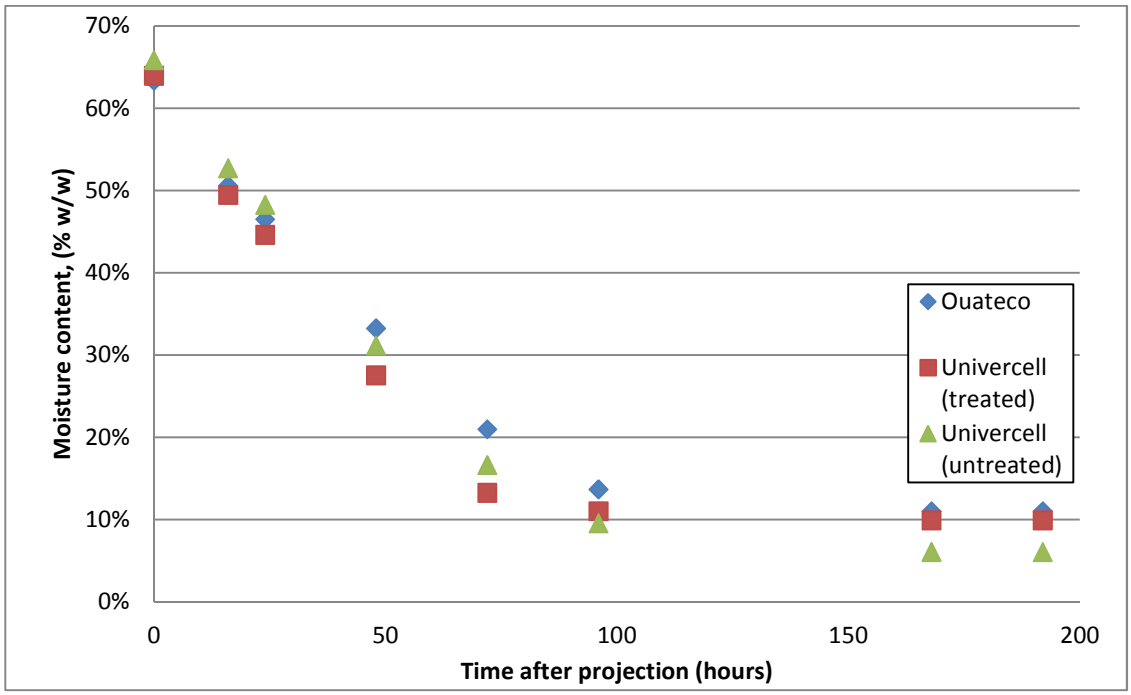


Figure 2.9: Drying of cellulose samples, comparison of different source material and treatment.

2.2.8.2 Influence of installed water dosage on drying

The drying curves of projected 300x300x90mm^3 samples with varying water pressure are shown in Figure 2.10. All samples show an initial linear constant drying rate, which corresponds to the removal of free water, and around 25% moisture content, the drying rate slow down until the material reaches equilibrium moisture content. This point of inflection is close to the point in which only bound water is present in the fibres, as is shown in Figure 2.6, which could mean that below 25% mainly bound water is present, which is harder to remove from the fibres, hence the slower drying rate.

Evidently, an increase in water dosage increases the time for the material to reach an equilibrium dry state. In practice, installers of cellulose fibre insulation consider that when the material reaches around 20% moisture content, it is considered dry enough to allow the installation of a vapour barrier and drywall. For example, a water dosage of 70% can increase the time to reach 20% moisture to 58 hours versus the 19 hours needed

when the minimum recommended dosage of 40% is applied, over three times the standard waiting time. It is worth noting that this applies only to 90mm thick insulation at the constant conditions of 25°C at 60% RH. In reality, the drying rates vary depending on thickness and ambient conditions. The thicker the specimen, the colder and more humid drying conditions are, the longer the drying times, which sometimes cause unacceptable delays in a construction project.

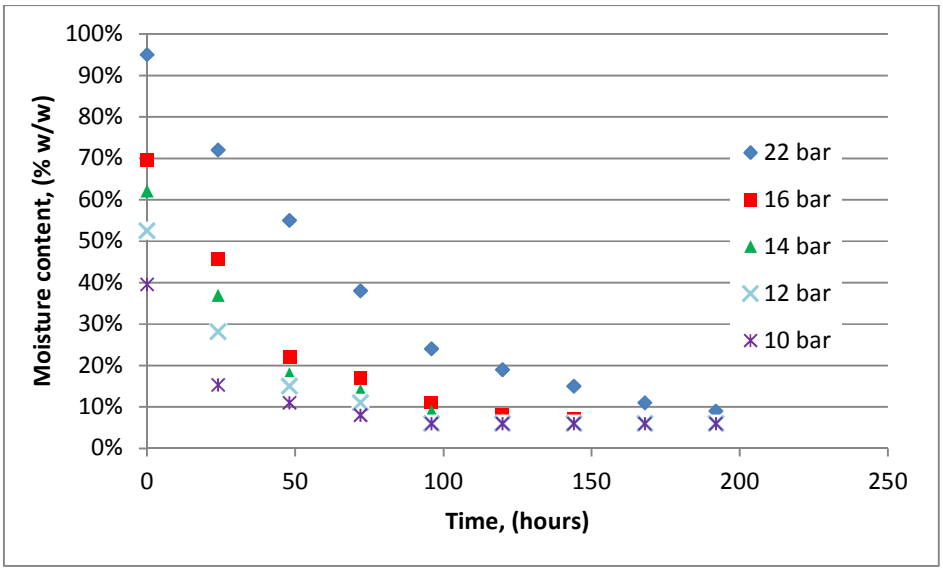


Figure 2.10: Drying of cellulose samples, with varying installed water pressure.

2.2.9 Changes in density

As shown on Figure 2.11, the final density of the material depends on the installed moisture content. The values shown are slightly higher than those found by Salonvaara et al. (2010). The first point in the series corresponds to the initial moisture content and density before spraying (blown density). At the minimum moisture content of around 40%, density varies from 47 to 53 kg/m^3. The results show a semi linear increase of density with initial moisture content up to 77% moisture. Past this point the density increase is at a much slower rate. When cross referencing this inflection point with Figure 2.5, one could observe that at the 77% moisture point almost all of the bound water has been absorbed within the fibres, while free water content continues to increase. The changes in density has two probable sources: the increased pressure from the pulverised water, which causes compaction of the material as it is projected, and the strengthening

and hardening of fibres as they dry. It could be feasible that the free water within the fibres contribute less to the increase in density than the bound water. As it has been shown with other natural building materials (Elfordy et al., 2008), these variations in density can also cause changes in the thermal and mechanical properties of the material, as will be shown in the following sections. It is therefore necessary not only to observe the influence of the sprayed moisture content on the mechanical and thermal properties, but to differentiate it from the influence of the increase in density induced by sprayed water. This further emphasises the need to control of water dosage since as the density increases, more of the material is used to insulate the same volume, which subsequently increases costs of installing the insulation.

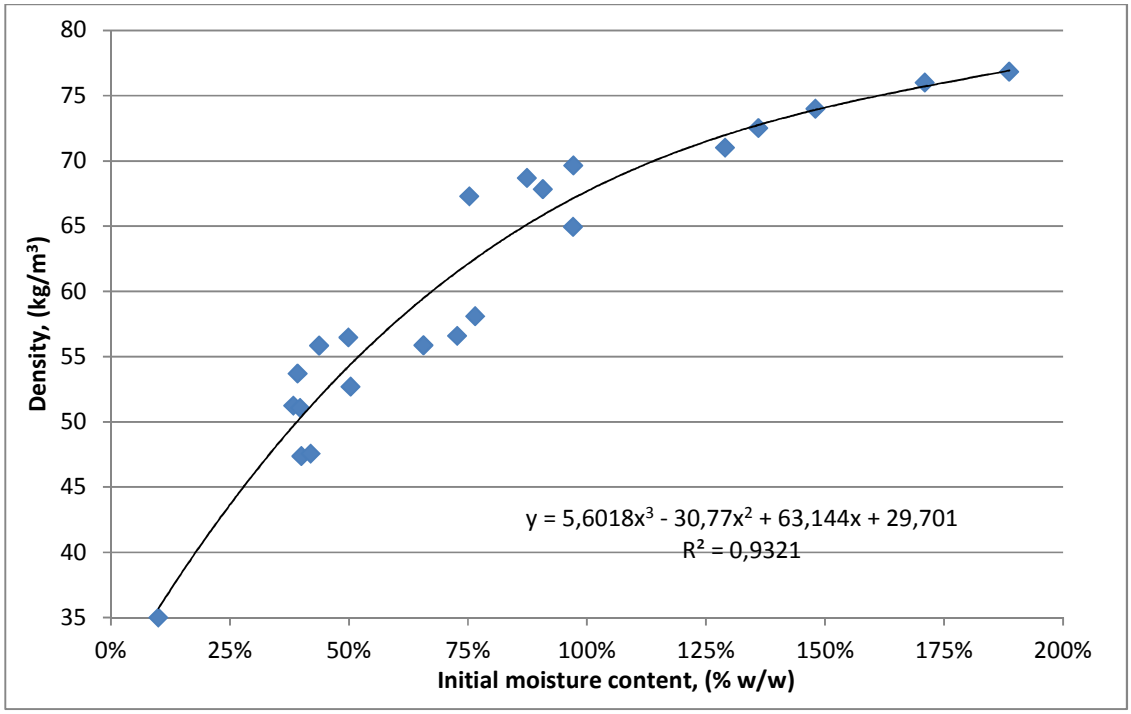


Figure 2.11: Influence of installed moisture on final density at equilibrium, with polynomial regression line.

2.2.10 Mechanical testing

Up until now there is no defined method to characterize the mechanical behaviour of wet spray CFI. The closest analogue would be the European standard EN 1605 and 1607 which define compression and tensile tests for insulation materials, but this is mainly applied to rigid, prefabricated insulation boards. Using the EN1605 standard as a

base, a compression test was developed to characterize wet spray CFI. The method is described in detail in EM11.

Stress-strain curves of 100x100 mm^2 cellulose insulation samples (see Figure 2.12) with different moisture dosages are shown in Figure 2.13. The lightweight material shows a quasi-linear elastic behaviour with no rupture at strains up to 20%. At higher deformation levels, the material starts to consolidate and the slope of the stress strain curve increases. As was expected, the material shows an increase in its mechanical resistance with increasing moisture content.



Figure 2.12: Compression test sample.

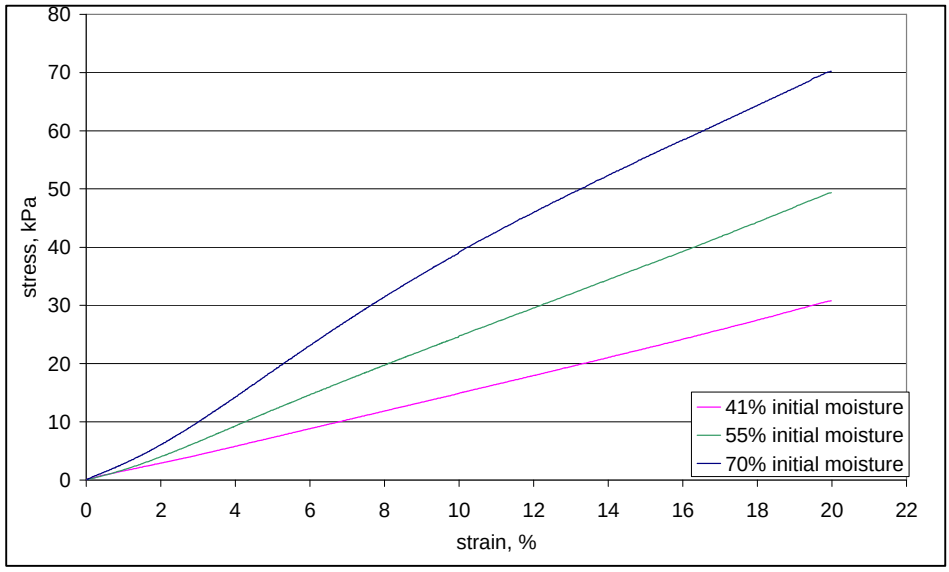


Figure 2.13: Stress-strain curves of projected cellulose insulation samples with varying moisture content.

Figures 2.14 and 2.15 show the relation between the moisture content, the calculated elasticity modulus E, and the calculated stress at 5% and 10% strain ($\sigma_{5\%}$ $\sigma_{10\%}$) respectively. All values indicate an increase with moisture content. As the material is compacted, the heterogeneous voids within it are filled with fibres, which could translate to an increase in mechanical resistance.

In order to define a minimum mechanical threshold of wet spray cellulose insulation that ensures the material will not sag or tear once projected, the measured values of the modulus of elasticity E and compressive stresses $\sigma_{5\%}$ $\sigma_{10\%}$ at around 40% moisture content are averaged. This gives an average of 14.05 kPa, 0.62 kPa, and 1.34 kPa for E, $\sigma_{5\%}$, and $\sigma_{10\%}$ respectively. These values could potentially be used as a reference point if, for example, a raw material (recycled newsprint) of different quality is used, or if the amount and/or type of additives are changed. Below 40% the CFI samples could either not be sprayed or crumble under their own weight.

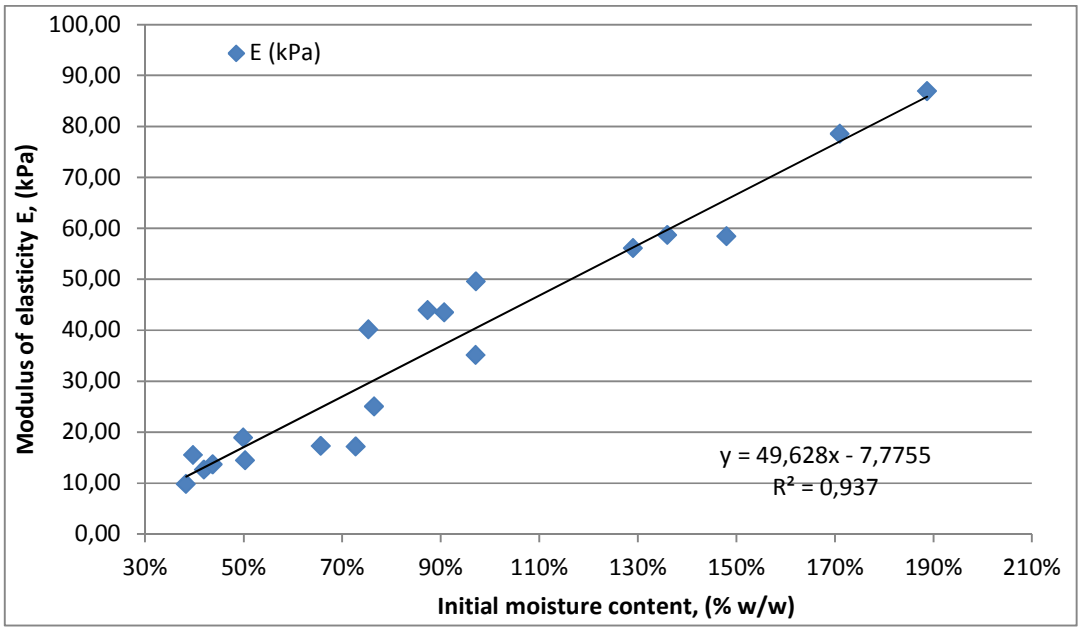


Figure 2.14: Influence of installed moisture content on modulus of elasticity E, with linear regression line.

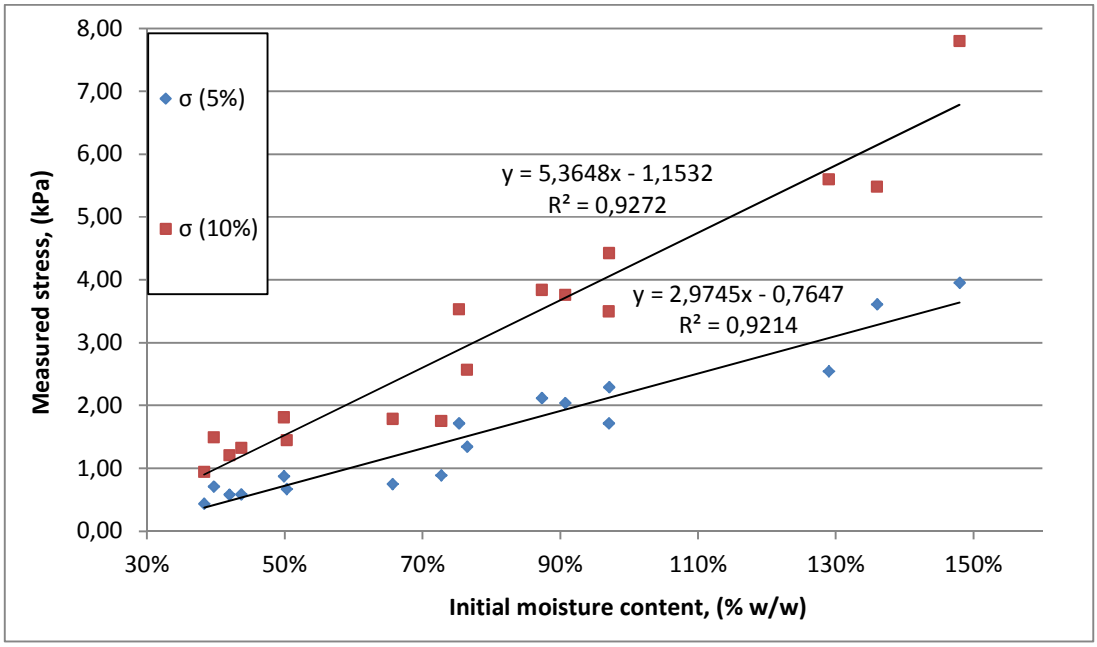


Figure 2.15: Influence of installed moisture content on $\sigma_{5\%}$ $\sigma_{10\%,}$ with linear regression line.

In order to account for the influence on the direct compaction of the material by the sprayed water, the previous results were also plotted with dry density of the samples, compared with samples sprayed at the minimum 40% moisture content and compacted

once dry, to artificially increase density. The results for modulus E and measured stresses are shown in Figures 2.16 and 2.17.

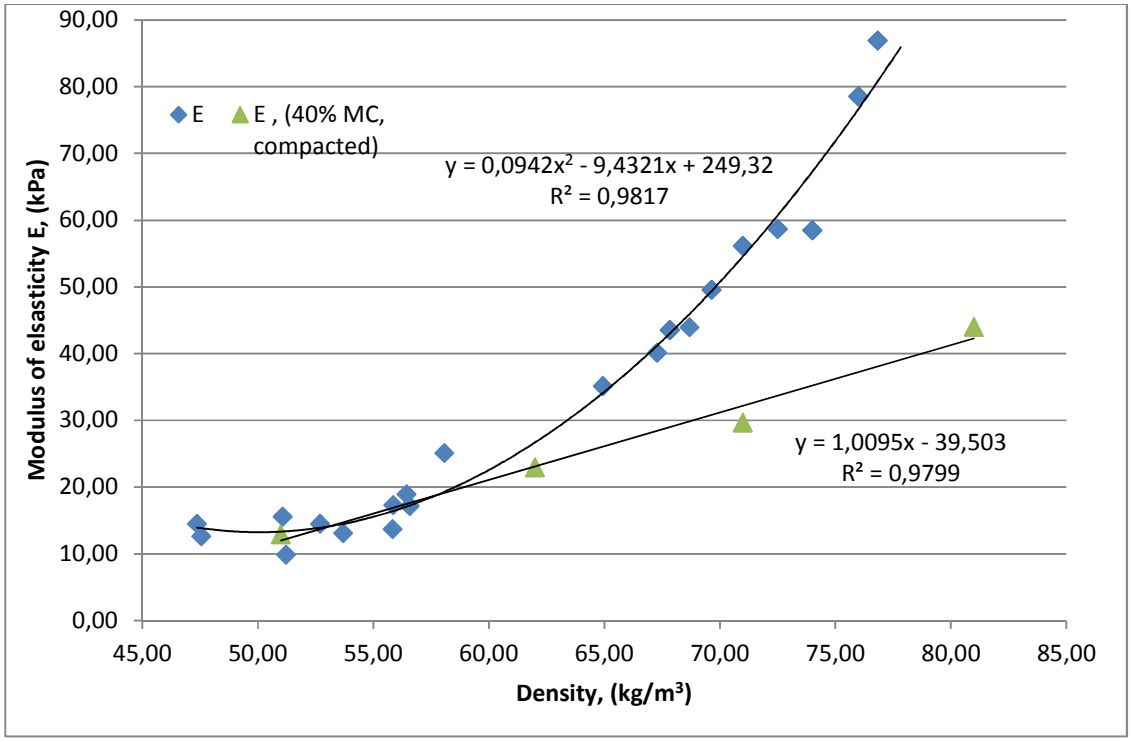


Figure 2.16: Influence of final density after spraying with various water dosages on modulus of elasticity E, compared to compacted samples, with regression lines.

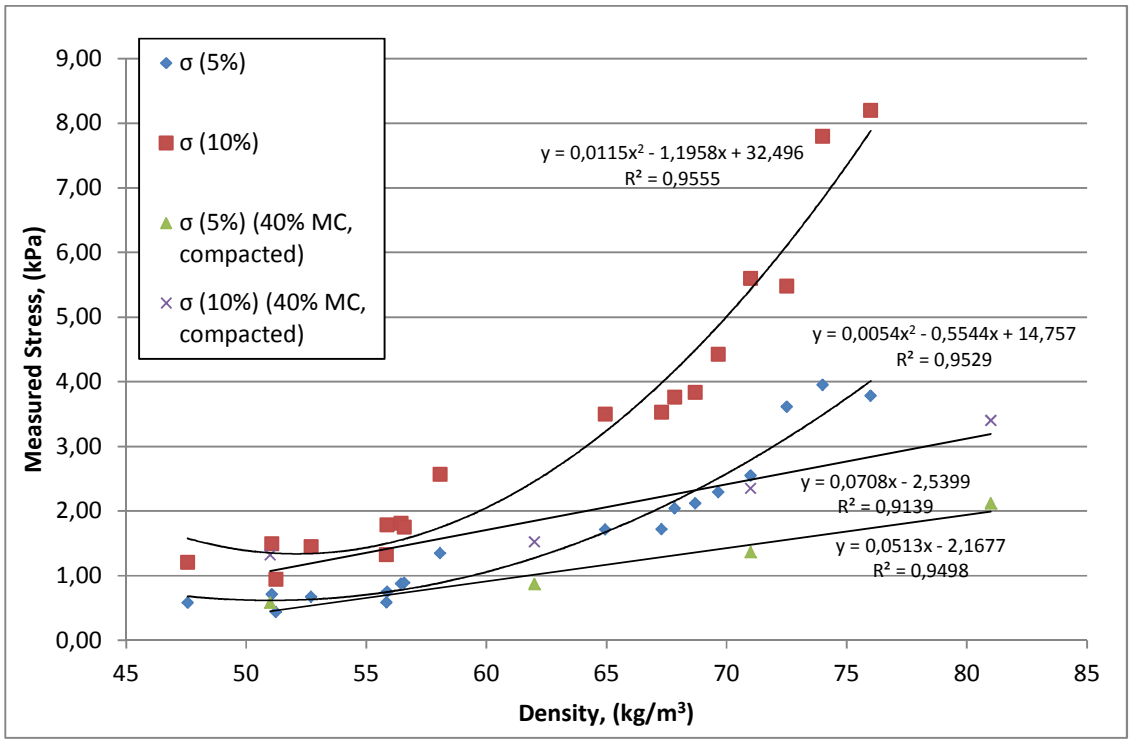


Figure 2.17: Influence of final density after spraying with various water dosages on $\sigma_{5\%}$ $\sigma_{10\%}$, compared to compacted samples, with regression lines.

When measured with regards to density, the mechanical resistance of the material behaves differently at different densities. For the typical "sprayed" densities, around 45-55 kg/m^3, the mechanical resistance increases slightly and its behaviour is similar to that of cellulose that has been dry compacted. Past a certain density, the mechanical resistance increases greatly, at a slope much higher than that of the compacted reference. This confirms the statement that the added sprayed water not only compacts the material, making it resist settling and fissures, but the swelling and hardening from the fibres as they dry has an important impact on the mechanical performance of wet spray cellulose insulation. This circumstance supports the case for using an additive, which could increase its mechanical strength while maintaining the low density of the material.

2.2.11 Thermal conductivity testing

The value of thermal conductivity, λ measured in W/mK is the main defining characteristic of a thermal insulation material. The lower the value of λ for a given material, the better it is as an insulator. Thermal conductivity measurements were applied

on 150x150x50mm^3 samples according to EM12. Figure 2.18 shows the variation of dry thermal conductivity with installed water dosage at average temperatures of 10°C and 25°C (λ_{10} and λ_{25}). As with mechanical properties, the value of dry thermal conductivity increased with installed water content. In this case the difference in values is much smaller, making the results more prone to irregularity. Thermal conductivity values varied from 37 to 43 mW/m.K, for measurements made at 10°C and around 40 to 46 mW/m.K for 25°C. These values are comparable to traditional insulation materials such as mineral wool. The high scatter could be due to the heterogeneity of the samples, or to irregularities in the contact surface between the heating plate and the sample. Since the material is highly compressible it was difficult to ensure perfect contact between the plates and the samples without compressing the samples, which would increase its density and thermal conductivity. While this increase in values of λ with applied moisture content is slight, it is still unfavourable to a wall's thermal performance. It is in the cellulose installers' best interest to use the lowest feasible water dosage in order to ensure optimum insulation capacity of the material.

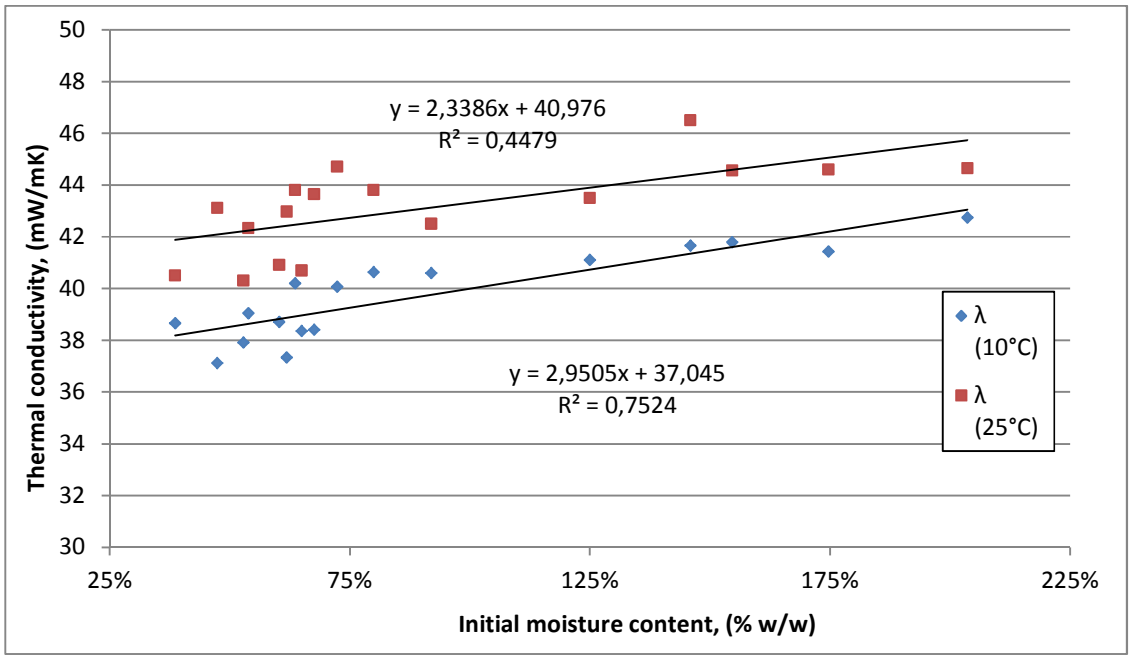


Figure 2.18: Influence of installed moisture content on thermal conductivity values λ_{10} and λ_{25}, with linear regression lines.

The influence of density of samples with varying spray dosage, is shown on Figure 2.19. The values are compared with samples compacted sprayed at minimum dosage once they were dry. Ideally the relation with density should give a clearer indication on the trend of thermal conductivity, but results show a high scatter as well, due to the closeness between values and the measurement issues mentioned previously. Nevertheless, in general terms, an increase was observed with thermal conductivity and as a function of density of the material. As it was found with compression tests, a slightly higher thermal conductivity was found from the sprayed samples than with the dry compacted samples, for samples with similar density. This could be due to the decrease in porosity from the swelling and drying of fibres, which differ from the decrease in porosity from compaction. The voids formed from these changes in porosity directly impact the thermal conductivity of the material.

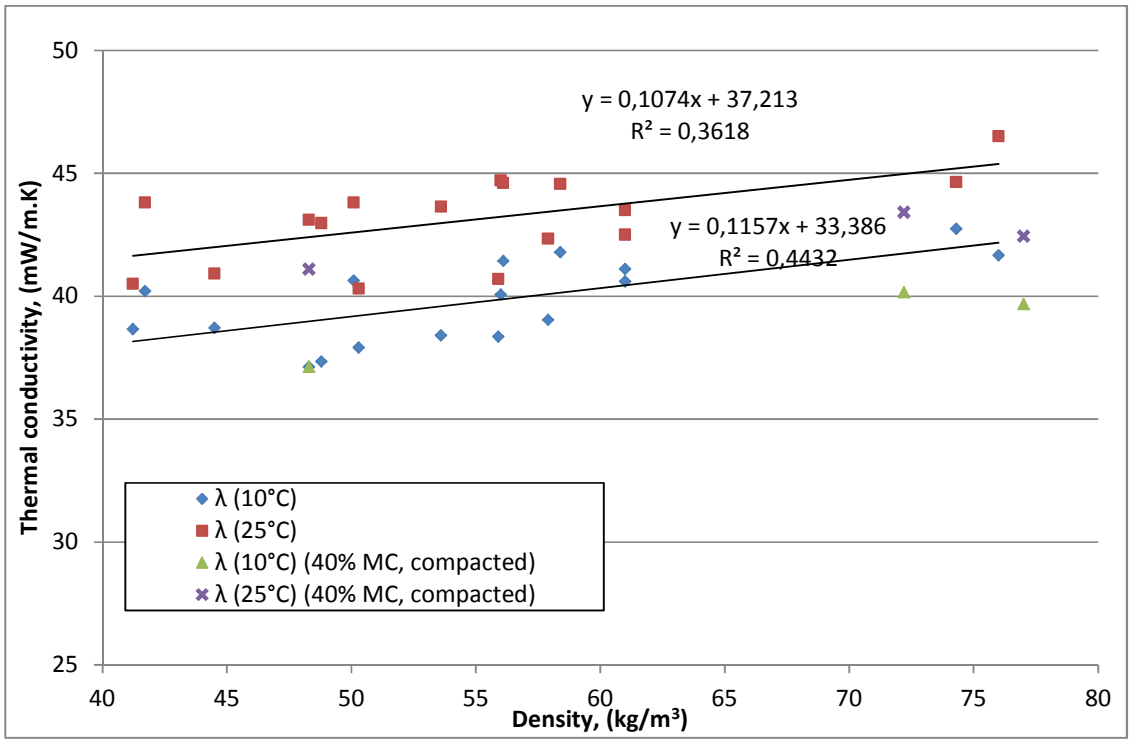


Figure 2.19: Influence of dry density on thermal conductivity values λ_{10} and $\lambda_{25,}$ with linear regression lines.

2.3 CONCLUSIONS

Many factors within the properties of cellulose insulation fibres can contribute to their behaviour with water. Of the two brands of cellulose insulation samples that were tested both showed some differences in these properties which, as was shown, translated into differences in drying performance of wet sprayed cellulose samples. These properties can include its chemistry and proportions of lignin, cellulose and hemicelluloses, the particle size distribution and morphology of the fibres, and the included additives. Other forms of characterization of cellulosic fibres and their relation with water include water retention, dynamic vapour sorption, and differential scanning calorimetry to determine bound and unbound water content. Unfortunately due to the variability in the raw material, due to the differences of the recycled newspaper, these factors are difficult to control and optimize. Nevertheless they give indications on the behaviour of the fibres with water and their subsequent drying once applied.

As the results have shown, the increase of water not only delays construction after installation, but also increases the density and thermal conductivity wet sprayed cellulose insulation. The increase in density and drying time with increased water dosage remain important factors to consider when applying cellulose insulation. A method to determine the mechanical behaviour of wet sprayed cellulose was designed. While in reality the compression tests don't fully represent the behaviour of cellulose fibres once they are projected, they do give an indication of how the installed moisture content strengthens the material in order to prevent sagging or tearing of the material. A baseline of 14.05 kPa modulus of elasticity E was defined as a minimum mechanical property of the material (at ambient humidity conditions) to prevent sagging. It was found that the applied water not only densifies the material, but as the fibres swell and become rigid during drying, an increase in the mechanical performance can be observed. In a practical sense, it would be pertinent to have a quality control system which the wet spray water dosage was measured in a test sample before applying to an entire wall. Mechanical tests such as the one developed in this work could help verify that with the proper applied water dosage, the material can maintain a minimum mechanical resistance to prevent settling. While the changes in thermal conductivity could be considered insignificant, there is still a loss in thermal efficiency of the material once an excessive amount of water has been used.

These results will serve as a basis to better understand the behaviour of cellulose insulation with added additives which improve its drying time and mechanical properties.

3

BIOBASED ADDITIVES AND THEIR INCORPORATION WITH WET SPRAY CELLULOSE INSULATION

3.1 INTRODUCTION

Having established the properties of cellulose insulation, as well as its behaviour with water using the wet-spray application process in previous sections, work could commence on the search for a proper additive which improves its performance in terms of drying. The information gained from the state of the art, detailed in chapter one states the issues associated with wet spray cellulose which could be improved via the incorporation of additives. It also catalogued and defined the viable biobased additives which could be used with cellulose insulation. The experimental results from chapter two further detailed the wet spray process and the influence of water on the final properties of cellulose insulation. It was found that increasing initial water content increased the density, mechanical strength and thermal conductivity of cellulose insulation after drying. While it's important that the material should have enough sprayed water to provide sufficient resistance to settling and tearing, an increase in density (which in turn increases its price since more material is used to insulate the same volume) and an increase in thermal conductivity decrease its interest as an insulation material. It is therefore

necessary to make the compromise between applying enough water to provide good cohesion within the material, and limiting the water dosage as to not have high during times and negatively affect the insulation material's properties. The same logic could be applied to the change in formulation. Ideally, the included additive should have little effect on the density and thermal conductivity of the material, while still decreasing the time required to reach a dry state and reinforcing its resistance to settling.

The mechanisms in which biobased additives could improve drying times are numerous: one strategy could be through the evaporation of water from the included additive as it hardens, which competes with the drying of the cellulose fibres, making them dry faster. Another could be the reduction of initial added moisture, using the mixed additive to compensate for the reduced water dosage to reinforce the material. In the previous chapter, a minimal mechanical strength attributed to the minimal recommended water dosage of 40% was determined. It could therefore be plausible to apply the material below this dosage, with an added binder which would help attain this minimal resistance. Another simple solution could be the increase in the materials initial dry matter content by using the additive to increase the solids ratio of the sprayed solution to make the material have lower initial moisture. Finally, the decrease in surface tension of the projected water via the use of surfactants could potentially make water evaporate from the material faster.

One of the limiting factors of the included additive could be its influence on the viscosity of sprayed solution. The ideal formulation should be applied with the standard spraying equipment, which is suited for liquids with water-like viscosity. Highly viscous liquids could clog the equipment or restrict the flow of the sprayed water, especially the nozzles of the spray hose. Consequently, especially in the case of binders, the additive/water proportions must be low enough as to not affect these factors, while still being effective in improving cohesion and drying times. This is another factor to consider as well as with the potential influence of the additive concentration on the materials final density and thermal conductivity.

The work in this chapter seeks to consider these factors in the search for a new formulation in wet spray cellulose fibre insulation with improved drying properties. Firstly, the feasible candidate biobased additives presented in the literature are studied

with regards to their viscosity and surface tension according to their concentration in aqueous solution. Their compatibility with the spraying material, the pump and spraying nozzles are then assessed. Finally, new formulations of wet spray cellulose insulation with biobased additives are characterized with regards to their drying times, density, compression resistance, and thermal conductivity. In summary, a Venn diagram which exemplifies the compromise of the properties of the ideal additive to be incorporated with cellulose fibre insulation is presented in Figure 3.1. The ideal additive must: improve the drying properties of cellulose, while still being usable with the spraying equipment and not hinder the properties of the final product.

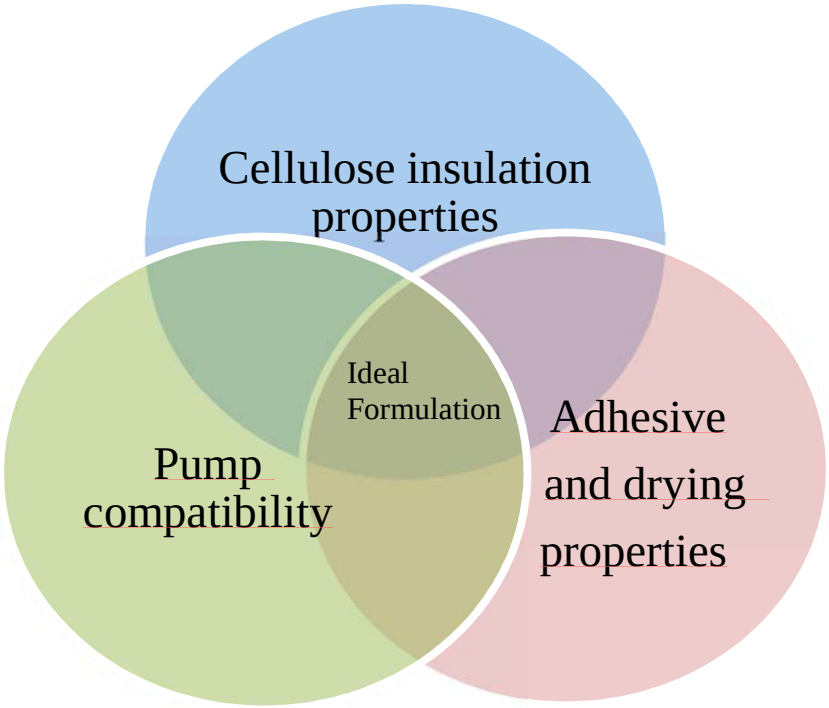


Figure 3.1: Optimal additive formulation Venn diagram.

3.2 BIOBASED ADHESIVES

3.2.1 Formulation and preparation

Biobased binders usually come in a powdered form which is then dissolved in aqueous solution to obtain the desired adhesive strength. The method in which the biobased adhesives are incorporated with the cellulose insulation needed to be evaluated. There exist two strategies in which this adhesive could be mixed with the cellulose

insulation. The powdered adhesive could either be dry mixed with the fibres before projection, just as is the case with the fire resistance additives, or they could be mixed with the sprayed water before projection.

Since water is added after the adhesives are incorporated within the fibres, liquid viscosity is not an issue with the dry mixing method, and thus the technique allows for an improved control of the dosage of the powder on the fibres. Despite these benefits, the high volume of the cellulose fibres would require either the use of high volume specialized mixing equipment before installation, or for it to be installed during manufacture. Another issue is the fact that these binders usually require to be fully dissolved in water to activate, and the added water during the spray application (40%-60% the mass of cellulose insulation) might not be enough, especially since the hydration of the binder competes with the moisture sorption of the fibres. This phenomenon is documented in the case of hemp concrete, where the lack of water within the binder due to moisture sorption of the hemp fibres hinder the adhesion of the binder to the fibres, making the material agglomerate and crumble (Amziane and Arnaud, 2013).

The wet process presents a different set of benefits and challenges. The powdered additive could be easily mixed onsite before spraying, allowing a better hydration than the dry method. However, most of the binders cited in the state of the art increase the viscosity of its solvent. This increase in viscosity could be problematic in the spraying process, where if the liquid is too viscous it might block the pump membrane or the spray tips.

Given these properties, it has been decided that the wet mixing method is better suited for the wet spray process. While the liquids viscosity is an issue, the concentration of the binder could be studied in order to attain a sprayable viscosity.

As the binder concentration in water decreases, its viscosity decreases, but also its effectiveness as a binder. These properties could also be modified using heat or varying pH., but this is not viable onsite during the spraying process. For example, roughly 1 tonne of cellulose insulation is needed to insulate one house. If one is to assume 40% added moisture, 400kg of water would be needed. It would then be cost prohibitive for a building project to either heat up or change the acidity of such a high volume of water. However, for some binders a pre-solution with a different pH levels could be made before

mixing with the final spraying water. The additives chosen for screening included the following: gums (guar gum and xanthan gum), polysaccharides (citrus pectin, pregelatinized corn starch, maltodextrin, and sodium alginate), a soy protein isolate, phenolics (lignosulfonate and low sulfonate lignin), and a reference polyvinyl acetate powder (PVAc). Since it has been shown that paper pulp has a negative charge, the use of cationic adhesives, which would be favourable for the retention of binders, was also explored. These include cationic tannin, chitosan, and cationic starch. The origin and preparation of these binders is detailed in EM 13.

3.2.2 Viscosity of biobased adhesive solutions

In rheological terms, biobased polymers are viscoelastic materials, which means that they some of the elastic properties of an ideal solid and some of the flow properties of an ideal liquid. The apparent viscosity of a liquid is a measurement which quantifies its intrinsic resistance to flow. It is measured through the determination of its resistance to gradual deformation by shear or tensile stress. It is usually measured in Pascals seconds (Pa.s) or Centipoise (cp). It is important to distinguish liquids which present Newtonian flow, i.e. the viscosity is independent of the shear stress, and non-Newtonian, in which viscosity can either increase or decreases with increasing shear stress.

Viscosity can directly affect liquid flow through a vessel such as a pump or hose. It is therefore necessary to determine the concentration – viscosity relation of each binder in order to find a maximal sprayed concentration that is compatible with the water pump and spray nozzle. Neither the water pump nor the spraying nozzles give indications on the maximum allowable viscosity so this needs to be determined practically. An initial test was done to determine the maximum viscosity that is compatible with the pumping and spraying equipment (see EM 13).

For most natural polymers, one can distinguish two phases of the concentration – viscosity relation. For highly diluted polymers, molecules are allowed to move freely. As concentration increases, the molecules start overlapping and forming an entanglement network, in which the slope of the viscosity increases. The point at when this occurs is known as the critical overlap concentration C*, as can be exemplified in Figure 3.2. Below this viscosity, the liquids exhibit a Newtonian steady-shear flow (Sworn, 2007).

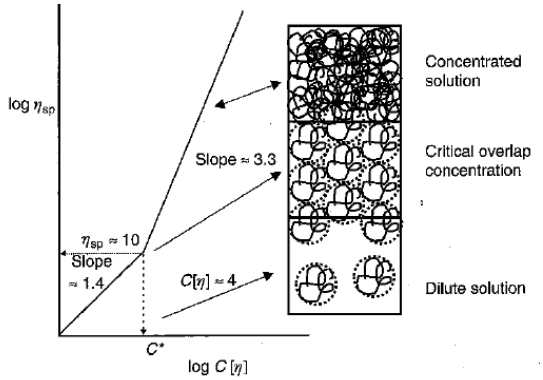


Figure 3.2: Concentration dependence of viscosity for polysaccharides, from (Sworn, 2007).

Viscosities of binders at different concentrations were tested according to EM14. Most of the tested concentrations showed a rheological behaviour similar to that shown for pectin, in Figure 3.3. For most concentrations, the viscosity is clearly independent of the shear rate; characteristic of a Newtonian or semi-Newtonian flow. For these cases, a shear rate of $20s^{-1}$ was considered to be the threshold for sprayed viscosity, since beyond this level, viscosities didn't vary. In others, as is the case of pectins at 10% concentration, non-Newtonian flow was noticed.

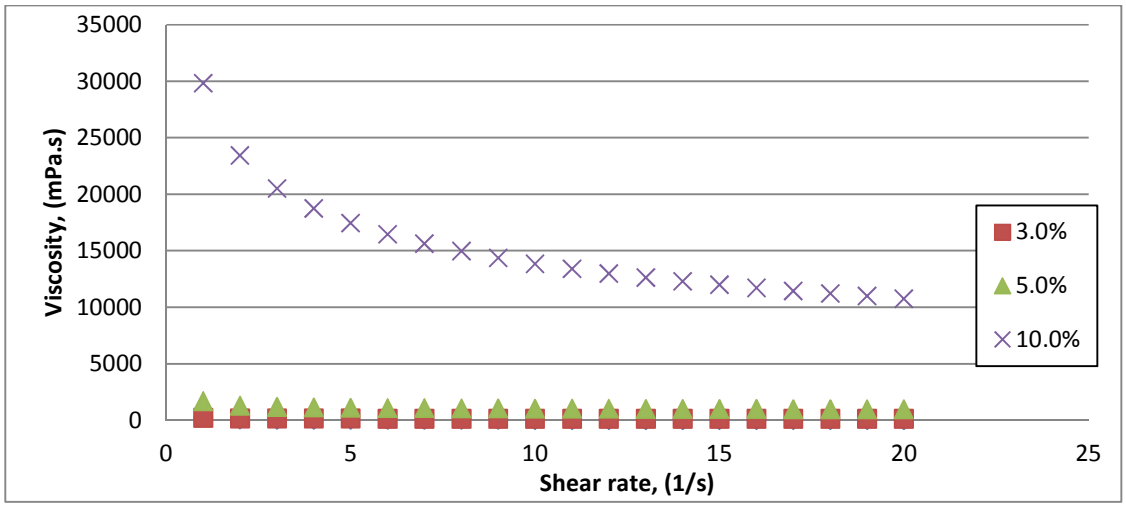


Figure 3.3: Viscosity-shear curve of pectin at varying concentration.

This could be an interesting factor to consider in practice with the spraying equipment, where if a concentration is "unpumpable" a higher flow rate through the

pump mechanism could dynamically increase the shear rate of the liquid passing through the pump hose and spray nozzles, thus decreasing the viscosity to a “pumpable” level. In these cases the viscosity needs to be re-tested at a higher shear rate. Although the specific shear rate of the pump and spraying mechanism are not known and are dependent on the applied pump rate, shear rates ranges have been detailed in (Gösta, 2003):

Process:	shear rate range:
stirring	$10^{1} – 10^{3}\ s^{-1}$
pumping	$10^{2} – 10^{3}\ s^{-1}$
spraying	$10^{3} – 10^{4}\ s^{-1}$

Table 3.1: Typical shear rates for different processes.

In practice, the liquid used for cellulose insulation is sheared as it is mixed, suctioned through the reservoir, pumped through a hose and sprayed through the nozzles. A shear rate of 3000 s^{-1}, corresponding to the range of shear induced through spaying was therefore tested for non-Newtonian concentrations which could feasibly have a low enough viscosity at high shear.

Results of the concentration-viscosity relation with different binders are shown in logarithmic scale in Figure 3.4. All binders show a wide range of concentration-dependant viscosities. All curves show a linear logarithmic behaviour, where some show a critical concentration point indicating an increase of the slope of the viscosity. Guar gum, xanthan gum and sodium alginate showed the highest viscosities, even at very low concentrations, while tannin, both lignin polymers, soy protein isolate, and maltodextrins’ viscosity increased just slightly at high concentrations. The viscosity of soy protein isolate increased at a higher pH due to the improved solubility of the solution. It could be possible that for some formulations the critical concentration was not within the range of concentrations tested. From the preliminary tests, with the spraying equipment, the maximum sprayable concentrations of 1% starch and 1.5% pectin correspond to viscosities of 35.5 and 37.1 mPa.s, respectively. This would suggest that the maximal sprayable viscosity for all liquids to be sprayed with cellulose fibre insulation would be around 40 mPa.s at the high shear levels. This limit is shown as a dashed line in Figure 3.4. This allows the categorization of the equivalent maximal allowable concentrations for all binders.

For most binders, the concentration in which a viscosity under this limit was attained was very low, showing Newtonian behaviour. However for some of the formulations of binders such as pectins, guar gum, xanthan gum, cationic starch, sodium alginate, and chitosan, a non-Newtonian shear-thinning flow was observed, in which a high enough shear could possibly produce a viscosity lower than the limit. These formulations were re-tested to the high shear of 3000 s^{-1}. The viscosity-concentration curves for these formulations at high shear rates are shown in Figure 3.5, with the 40 mPa.s limit shown. It was found that, at a high shear, the concentrations of cationic starch, guar gum, xanthan gum and chitosan could be higher than initially predicted with the low shear test, since viscosities at high shear are lower than the established limit.

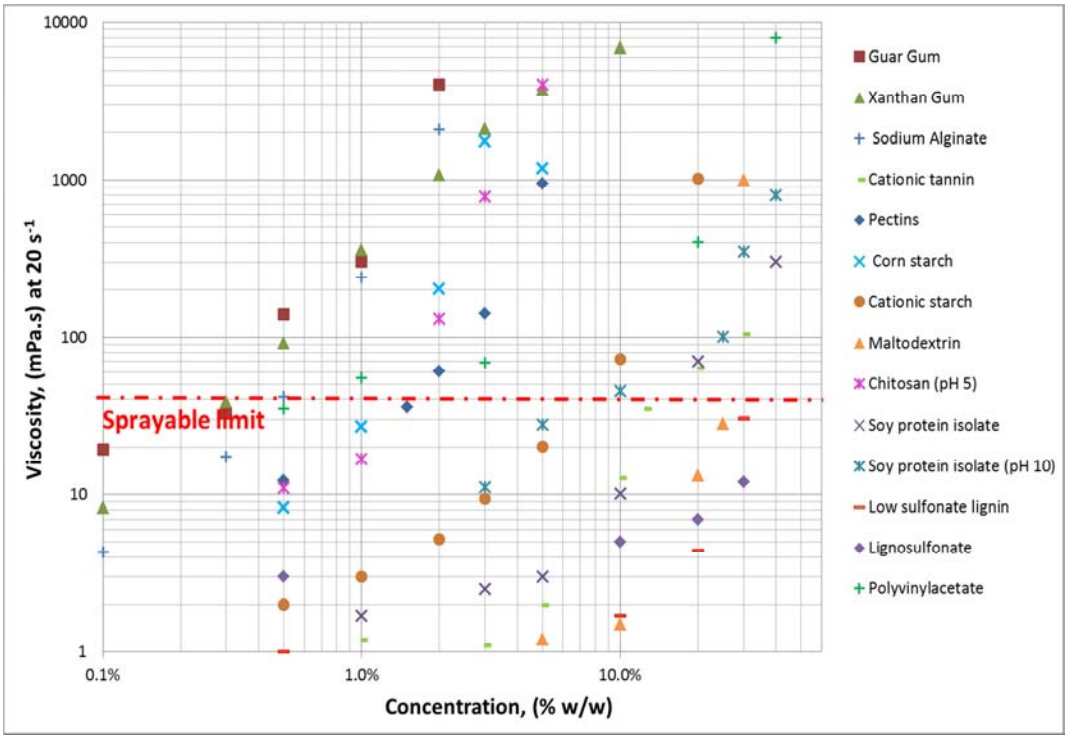


Figure 3.4: Viscosity concentration curves for binder solutions.

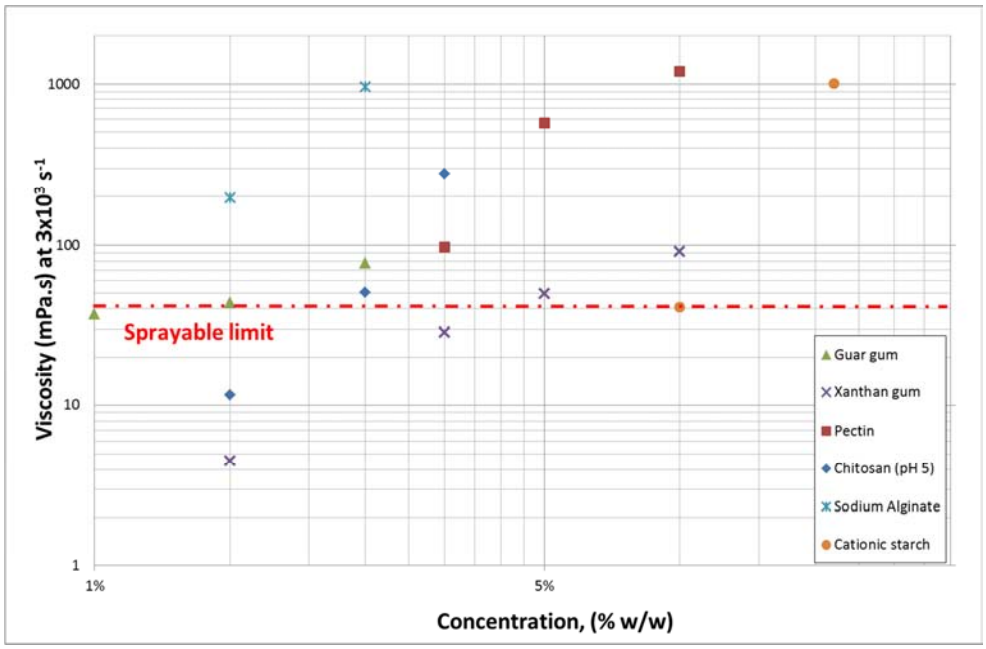


Figure 3.5: Viscosity concentration curves for shear thinning (non-Newtonian) binder solutions at high shear.

The possible sprayable concentrations for the biobased binders in both high and low shear conditions are presented in Tables 3.2 and 3.3. These need to be verified through direct testing with the pump and spray equipment. It is important to note that only applies to the specific pump and spraying mechanism used for this application. Although this equipment is the most commonly used for the spraying of wet spray cellulose insulation, others with different viscosity or shear rate limits could be used, but a study of alternative equipment exceeds the scope of this study. Another factor to consider is the fact that natural polysaccharide suspensions can degrade, losing its rheological and adhesive properties over time. It is an important issue to consider if the mixed liquid adhesive is stored onsite before application.

	Sprayable Concentration (% w/w)	Measured viscosity at 20 s^{-1} (mPa.s)
Pectin	1.5	36
Guar Gum	0.3	39
Xanthan Gum	0.3	32
Corn starch	1.0	27
Cationic starch	7.0	39
Maltodextrins	25.0	28
Sodium Alginate	0.5	42
Cationic tannin	12.5	35
Low sulfonate lignin	25.0	4
Lignosulfonate	25.0	7
Chitosan (pH= 5)	1.0	17
Soy protein isolate	15.0	40
Soy protein isolate (ph.=10)	5.0	28
Polyvinyl acetate	0.5	35

Table 3.2: Sprayable concentrations of binder solutions and viscosities.

	Sprayable concentration (Non-Newtonian) (% w/w)	Measured viscosity, 3000 s^{-1} (mPa.s)
Guar Gum	0.5	39
Xanthan Gum	3.0	28
Chitosan (pH= 5)	1.5	32
Cationic starch	10.0	41

Table 3.3: Sprayable concentrations of binder solutions and viscosities, at high shear rate.

3.2.3 Tensile strength

A compromise between the binders' viscosity and its effectiveness as a binder must be made. The ideal binder and concentration must have a minimal viscosity and maximal effective adhesion. In order to have an indication of the adhesion forces within the applied binders and their relation to water concentration, small scale tests must be made before testing with cellulose fibre insulation. A method to test the adhesion of newsprint paper strips with biobased binders was devised using tensile tests. The procedure is based on the work by Flory et al., (2013) and described in EM15. Small paper strips of the source newspaper for use with cellulose insulation were dipped in the

adhesive and subject to tensile force until rupture. The concentrations of biobased binders presented in viscosity experiments were tested with adhesive tensile tests in order to correlate the decrease of viscosity (favourable for projection with cellulose) with the decrease in adhesive forces (unfavourable for projection with cellulose). Results are presented in Figure 3.6 and compared with a reference (wetted and dried newspaper sample). As was predicted, binder concentration had an influence on the adhesive properties of paper reinforced with the binder. Adhesive performance varied on the type of binder. The tensile strength reference newsprint paper has an average value of 36.2 N, but also a high standard deviation of 5.2 N. Therefore, as presented in Figure 3.6, a range of "non adhesive" concentrations was defined for formulations which are within the range of this reference, with a limit threshold established at 36.2+5.2 = 41.4N.

For some binders, low concentrations had little or no effect to adhesion when compared to the reference paper sample. This indicates that they provide no advantage to using those adhesive formulations instead of just water. Maltodextrins and cationic starch showed no adhesion even at concentrations up to 25%. Corn starch and polyvinyl acetate showed high adhesion but for "pumpable" viscosities, they were not effective. Pectin, guar and xanthan gums displayed the highest tensile strength, but within the "sprayable" concentrations, measured adhesive strength was low yet still superior to that of the reference. As with viscosity, the increase of pH in soy protein isolates increases its adhesive strength.

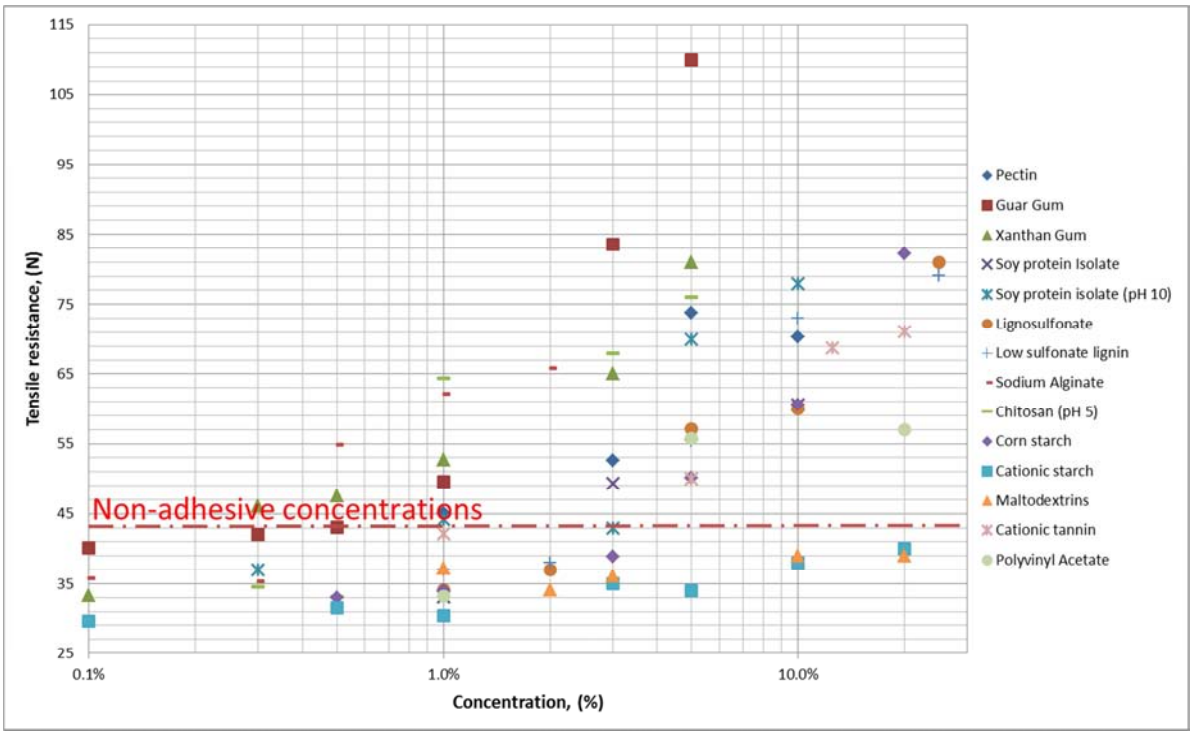


Figure 3.6: Tensile strength vs concentration of binder solutions.

It is possible then to cross-reference these results with those of the "sprayable" concentrations to determine which biobased binders would not be feasible to use with cellulose insulation, as is shown in Tables 3.4 and 3.5. Using the hypothesis that the liquid was sprayed at a high shear, these concentrations could be increased, producing better adhesion. This aids in refining the list of potential additives to test with the spraying equipment.

Additive	Sprayable concentration (% w/w)	Adhesive strength, (N)	Better than water?
Pectin	1.5	45	**Yes**
Guar Gum	0.3	42	**Yes**
Xanthan Gum	0.3	46	**Yes**
Corn starch	1.0	39	No
Cationic starch	7.0	34	No
Maltodextrins	25.0	36	No
Sodium Alginate	0.5	54	**Yes**
Cationic tannin	12.5	68	**Yes**
Soy protein isolate	15.0	60	**Yes**
Soy protein isolate (ph.=10)	5.0	70	**Yes**
Low sulfonate lignin	25	73	**Yes**
Lignosulfonate	25.0	79	**Yes**
Chitosan (pH= 5)	1.0	70	**Yes**
Polyvinyl acetate	0.5	36	No

Table 3.4: Comparison of tensile strength of binder solutions.

	Sprayable concentration (Non-Newtonian) (% w/w)	Adhesive strength, (N)	Better than water?
Guar Gum	0.5	43	**Yes**
Xanthan Gum	3.0	46	**Yes**
Chitosan (pH= 5)	1.5	70	**Yes**
Cationic starch	10.0	37	No

Table 3.5: Comparison of tensile strength of non-Newtonian binder solutions with high shear.

These binder concentrations are then theoretically usable with the spraying equipment and beneficial for the adhesion with cellulose insulation. It is worthwhile to note that the actual conditions in which cellulose is applied with these binders i.e. low liquid to cellulose ratio, makes it difficult to compare the mechanical performance of the final product with these tensile tests. In other words these small scale tensile tests are an indication of which formulations will not improve the mechanical resistance of cellulose insulation, and not necessarily which formulations actually will. Finally, in order to further narrow down the potential additives list, practical considerations were taken into account.

Since both gums showed similar adhesive and rheological properties in Newtonian concentrations, the more prevalent of the two, guar gum was considered. The same reasoning was applied in the case of low sulfonate lignin vs lignosulfonate, where lignosulfonate is the cheaper and more accessible of the two. Finally, since it would prove difficult to maintain the pH of 400L of the spray water, both binders which requires a buffer, Soy protein isolate (pH=10) and Chitosan (pH= 5) were excluded. The binder formulations to test for compatibility with cellulose insulation and its equipment are therefore:

Binder	Concentration (% w/w)
Pectin	1.5
Guar Gum	0.5 (non-Newtonian)
Soy protein isolate	15
Sodium Alginate	0.5
Cationic tannin	12.5
Lignosulfonate	25

Table 3.6: Final sprayable and adhesive concentrations.

3.2.4 Compatibility with spraying equipment

Once the optimal binders and concentrations were defined with regards to viscosity and adhesion performance, it is possible to test the additive formulations with the spraying and pumping equipment used in cellulose fibre insulation. The preliminary tests gave an indication of the pumpable concentrations for starch and pectin. These concentrations need to be confirmed for the other biobased binders. The binders must also be mixable and homogeneous at the scale in which cellulose insulation was applied. They also need to be able to be pumped and sprayed at the same flow rate as water in a typical application of wet spray cellulose. In order to verify these characteristics, the biobased adhesive powders were first mixed with 15L water at the determined concentrations using a Controlab 40L mixer on the lowest speed for 5 minutes. Once a homogeneous mixture was obtained, the liquid was poured on a bucket, covered and left to settle for another 10 minutes. Prior and after spraying, the pump, hoses and spray nozzles were rinsed by spraying small amounts of water. Small samples of the liquid were taken after mixing and their viscosity was measured using the same method described previously (EM14). The mixed liquid was then sprayed into buckets at different

pump pressures during 30 seconds and the flow rate was estimated by weighing the buckets after spraying. If little to no liquid was sprayed, the concentration was considered unsprayable. To further optimize the binders, a slightly higher concentration than the ones defined previously were also tested to ensure that the stated maximum could not be higher.

Initially during mixing, the guar gum mixtures showed instances of solids clumping, which blocked the flow through the spray nozzles. In order to have a homogeneous mixture without clumps, the solid clumps were filtered using a 5mm wire mesh, and mixed again with a small amount of the liquid adhesive.

Results of liquid flow for some formulations with the measured viscosities and different pump pressures are shown in Table 3.7. For the most part, the initial estimated pump limit viscosity of 40 mPa.s was valid. In some cases the maximum pump pressure of 24 bars could not be attained (indicated as “pump limit” in Table 3.7). This means that the increased viscosity of the pump stresses the pump motor making it pump at a higher pressure in the one displayed in the pressure gauge. This was more prevalent in the more viscous, non-Newtonian concentrations such as guar gum and sodium alginate. A pressure of 10 bar is used to attain the 40% moisture content minimum for cellulose insulation sprayed with water, with a flow rate of around 2 L/minute, consequently, a slightly higher pump pressure needs to be used to attain the same flow rate as in water. For sodium alginate, a higher maximum sprayable concentration of 0.75% could be utilized if the pump pressure was adjusted to around 15 bar. This increase in sprayable viscosity establishes the premise that the high shear induced from the spraying of shear thinning concentrations reduces their viscosity below the sprayable threshold. The fact that during mixing the liquid endures as a shear force could also be a factor. This can also be seen when spraying guar gum at 0.75% concentration, but the pump reaches its limit before it is possible to attain the required 2 L/minute flow rate.

Liquid	Concentration (% w/w)	Measured viscosity (mPa.s)	Spray rate (L/min) at applied pump pressure :					
			2 bar	6 bar	10 bar	14 bar	18 bar	24 bar
Water (reference)	100	1.00	0.84	1.48	2.01	2.23	2.89	3.04
Pectin	1.5	32.2	Not sprayable	1.42	1.92	2.14	2.46	2.81
Pectin	1.75	47.1	Not sprayable					
Guar gum	0.50	27.2	0.82	1.4	2.02	2.24	2.75	2.92
Guar gum	0.75	43.2	0.64	1.08	1.33	Pump limit	Pump limit	Pump limit
Sodium Alginate	0.5	22.4	0.8	1.38	2.00	2.2	2.77	3.01
Sodium Alginate	0.75	35.2	0.22	0.68	1.22	1.88	2.33	Pump limit
Sodium Alginate	1	52.3	Not sprayable					
Cationic Tannin	12.5	38.2	Not sprayable	1.24	1.82	2.05	Pump limit	Pump limit
Cationic Tannin	15	51.1	Not sprayable					
Lignosulfonate	25	11	0.84	1.48	2.01	2.23	2.89	3.04

Table 3.7: Pump rate of additives.

It is clear that viscosity is a limiting factor in the spraying of the adhesives. Some additives need to compensate with a higher sprayed pressure to attain the required flow, while others could not be sprayed at all.

3.3 SURFACTANTS

3.3.1 Formulation

Five different surfactants with varying properties were considered. Non-ionic surfactants with varying HLB (hydrophilic lipophilic balance) were tested. Tween 20, Triton X-100, and Poly(ethylene glycol) monooleate 400 (PEG-400). In order to test the influence of charge, cationic CTAB Cetyl trimethylammonium bromide, and anionic SDS (Sodium dodecyl sulphate) were also used. All surfactants were provided by Sigma-Aldrich. Their charge and HLB (hydrophilic lipophilic balance) values are summarized in

Table 3.8 (HLB can only be empirically calculated for non-ionic surfactants, meaning that the HLB for SDS and CTAB are comparative values).

	Type of surfactant	HLB Value
Triton X100	Non-ionic	13.51
Tween 20	Non-ionic	16.7
PEG-400	Non-ionic	12
CTAB	Cationic	10
SDS	Anionic	40

Table 3.8: Properties of surfactants.

Surfactants-water solutions show viscosities similar to that of water, which is beneficial for spraying. It is therefore unnecessary to measure its influence on viscosity. In order to determine the penetration capabilities of surfactant solutions through the voids and capillaries of cellulose insulation, surface tension measurements had to be performed.

3.3.2 Surface Tension

Surfactants could possibly improve drying conditions of cellulose through reduced surface tension of the liquid, making the liquid flow faster through the capillaries within the fibres. Surface tension measurements were made according to the method described in EM16. Concentrations of 0.1%; 0.5% 1% and 2% were tested.

	Surface tension (mN/m)			
Concentration (%w/w)	**0.10**	**0.5**	**1**	**2**
Triton X100	34	31	30	30
Tween 20	45	36	35	35
PEG-400	64	63	60	55
CTAB	40	37	37	38
SDS	31	31	31	31

Table 3.9: Surface tension measurements.

As expected, all surfactants lowered the surface tension of water (72 mN/m at 25°C), SDS and Triton provided the lowest surface tension. Past 0.5%, little change in the surface tension was noticed for all surfactants, except PEG, which requires a higher concentration to reach its minimum. For formulations with cellulose, a high concentration of 2% surfactant will be applied to ensure proper wetting of the fibres and surfactant adsorption.

3.3.3 Compatibility with spraying material

Since viscosity is not an issue with surfactants, the only concern with the use of surfactants is the production of foam during spraying. In fact if the water in the sprayed water reservoir has foam, this foam and not the liquid is suctioned through the pump, reducing the amount of liquid actually sprayed. This foam is mainly produced as the solution gets suctioned, passes through the pump and gets reintroduced to the reservoir via the discharge port. In order to prevent the suction of this foamed water, adjustments were made: first a higher amount of water was mixed in the reservoir to ensure a higher water to foam ratio, and next the suction hose was fastened on the bottom of the reservoir to ensure that only the water, which stays at the bottom was sprayed while the foam floats. Once these adjustments were made, surfactant formulations had the same flow rate as water. On the other hand, actually spraying foamed water with cellulose could prove beneficial since the foam might create voids within the material, possibly reducing its thermal conductivity. This is a technique often used in lightweight porous materials such as aerated autoclave concrete (AAC) (Schnitzler, 2006).

3.4 INFLUENCE OF ADDITIVES ON THE PROPERTIES OF CFI

3.4.2 Drying via binder evaporation and solids content increase

As was defined previously, the first strategy to reduce drying times for wet sprayed cellulose insulation with included additives was to apply a typical dosage of water with the included binder, having the drying of cellulose occur as the water evaporates from the binder, thus theoretically increasing the flow of moisture from the inside of the porous material to the surface. In order to test this theory, 300x300x100mm^3 cellulose samples were sprayed with the mixed adhesives via the same method used in Chapter 2 (EM10). After spraying, each sample had an initial liquid content of 40% ±3%. Samples were stored in the same ambient conditions (60% RH, 25°C), and their weight measured periodically. Only the viable formulations, compatible with the pump, hoses and nozzles, which were defined in the previous section, were tested. The drying curves of each formulation , compared to cellulose sprayed with water are shown in Figure 3.7,

The time to reach a dry state of 20% moisture content, using linear interpolation between points, is presented in Figure 3.8.

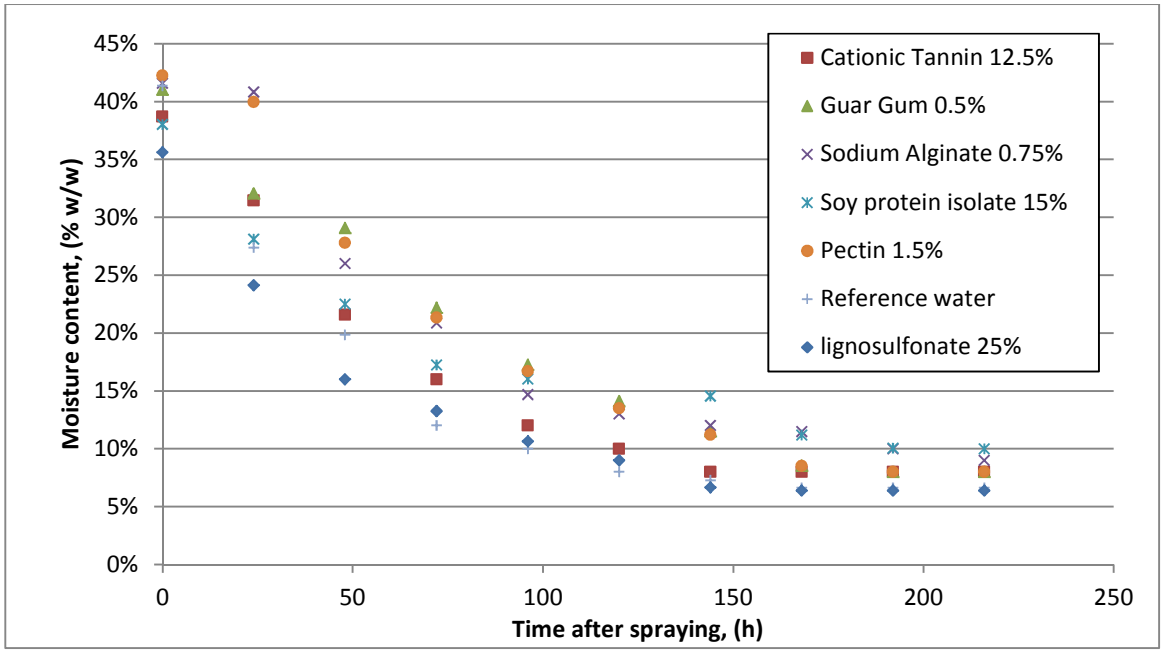


Figure 3.7: Drying curves for cellulose samples.

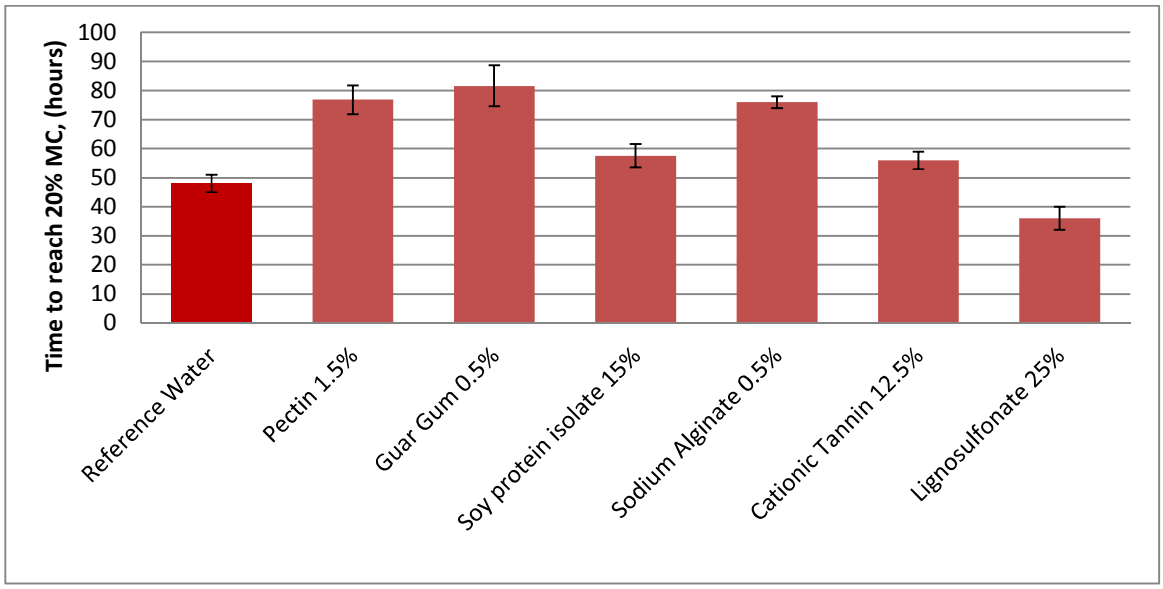


Figure 3.8: final sprayable and adhesive concentrations.

Results show that for most liquids sprayed at around 40% dosage, the contribution to the drying times by most of the available binders were either negligible or

detrimental to reach the dry state. The only exception was lignosulfonate, for which the dry state of 20% was reached 12 hours faster than the reference. This is partly due to the fact that, since a high concentration of lignosulfonates was used, the higher density liquid contained high solids content. The same volume of liquid was sprayed, but the liquid contained a 25% ratio of diluted solids, thus reducing the overall moisture content of the whole material, with an initial moisture content of 36%. This is also reflected with the other high concentration binders, although their drying slope during the first three days is slower than the reference.

There exist many explanations for these delays in drying times with biobased binder treated wet spray cellulose. One cause could be the formation of a “crust” on the surface of the cellulose; which is more exposed to the ambient air and humidity, thus hardens and crystallizes on the surface. This crust blocks moisture transport to the surface, making its drying slower. Similar results were found by Eloukabi et al., (2013), where efflorescence caused by dissolved minerals drying through a porous medium, created a crystallized surface which gives variations of the drying rate.

Another cause for this delay relates to the influence of viscosity on moisture flow through a porous medium. Darcy’s law is a relation which characterizes the relationship between the instant discharge rate of a fluid through a porous medium with the viscosity and moisture pressure rate, over a defined distance. The relation is as follows:

$$Q = \frac{-\kappa A(p_b - p_a)}{\mu L}$$

(3.1)

Where Q (m^3/s) is he total fluid flow rate, κ is the intrinsic permeability of the medium, (m^2), the cross-sectional area of the flow, A (m^2) and (p_b - p_a) (Pa), the total pressure drop, μ (Pa·s) is the viscosity of the liquid and (L) (m) is the length over which the pressure drop is taking place. When applied to the flow of a liquid through cellulose fibre insulation, the flow rate Q of a viscous liquid such as a binder decreases when compared to that of water due to two factors: first the decrease in the permeability (κ) of the cellulose fibres due to the crust formation, as mentioned beforehand, and the increase of μ due to the additives, which is inversely proportional to the flow rate Q, thus resulting in a reduced moisture flow through the cellulose insulation. In general the drying of a

building material occurs in two phases: a fast drying and a slow drying rate. The fast drying rate is generally attributed to the evaporation of the free (unbound) water from the large pores and capillaries of the material, while the slow drying period consists on the removal of the bound water.

Pectin and sodium alginate showed a slow initial drying period during the first 24 hours, which could be due to competition between the moisture sorption of the binders and the cellulose fibres. Some formulations also showed higher retention of moisture from the binders, with a higher final equilibrium moisture content of the cellulose insulation samples than the reference water sprayed samples.

As an interim conclusion, only lignosulfonates were found to be an interesting additive in reducing drying times for wet spray cellulose insulation using the increased solids content strategy. The next approach is to test surfactant solutions and additive – reinforced cellule insulation with lower sprayed moisture content.

3.4.3 Drying via surface tension reduction

The surfactants listed in section 3.4.1 were mixed with 2% concentration and sprayed with cellulose on the standard, 300x300x100mm^3 moulds. The initial moisture content was 40% ±3%. Table 3.9 indicates drying times to reach 20% moisture content, in comparison to the reference. All drying curves had the same shape with little variance with respect to the reference. Only the cationic surfactant (CTAB) had a significant improvement in drying times, drying 7 hours faster than the reference with only water.

Formulation	Time to reach 20% moisture content, (Hours)
Water Reference	51
Triton X100 2%	52
Tween 20 2%	51
PEG-400 2%	49
CTAB 2%	43
SDS 2%	48

Table 3.9: Drying times of surfactant samples.

This would seem to indicate that the actual reduction in surface tension of the sprayed water had little impact on the evaporation of moisture from the cellulose fibres while the ionic interactions between the cationic CTAB and the negatively charged cellulose fibres could provide a slight reduction in drying times. These results are parallel with the work of Beaupré (2012), where the surfactants with the lowest surface tension concentration did not necessarily produce the highest dewatering of cellulose pulp, and a high concentration of CTAB provided the best results.

3.4.4 Mechanical strength and density

The expectation in using biobased binders with wet sprayed cellulose insulation is that they can increase the mechanical performance of the material through the use of a smaller amount of water, or in the least maintain the base mechanical resistance at the minimum sprayed moisture dosage. Also it would be ideal if the additive would have less of an influence on density than sprayed water dosage while still maintaining or improving its mechanical performance. The dried samples sprayed at around 40% dosage from binder formulations described in 3.4.2 and CTAB sprayed samples from 3.4.3 were cut into nine 100x100x100mm^3 cubes as was done in chapter 2. Figures 3.9 and 3.10 show the measured elastic modulus, stress at 5% and 10% deformation respectively, with final density on the secondary axis. They are compared with 40% and 68% average moisture content reference standard wet sprayed cellulose insulation samples. Averages and standard deviation for measured values are represented, with 3 data points for density and 3x9=27 data points for mechanical resistance values.

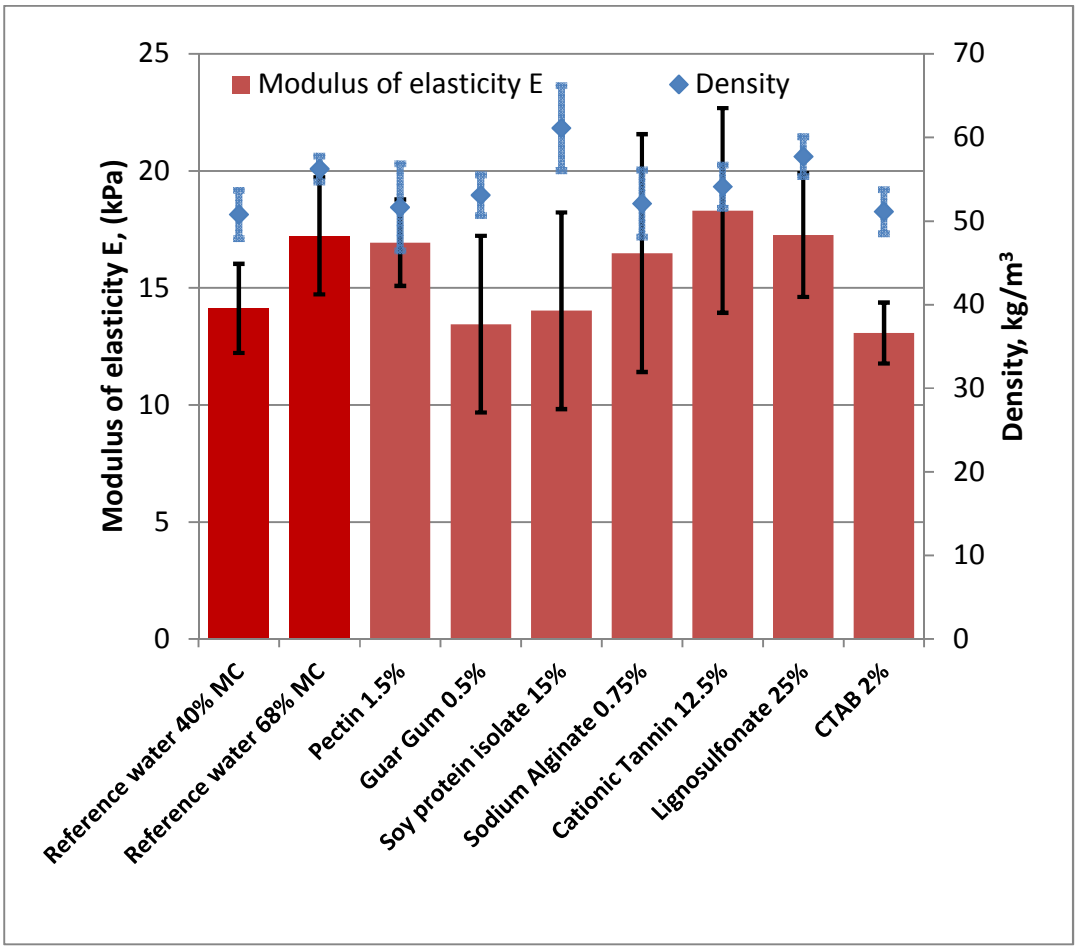


Figure 3.9: Modulus of elasticity of samples.

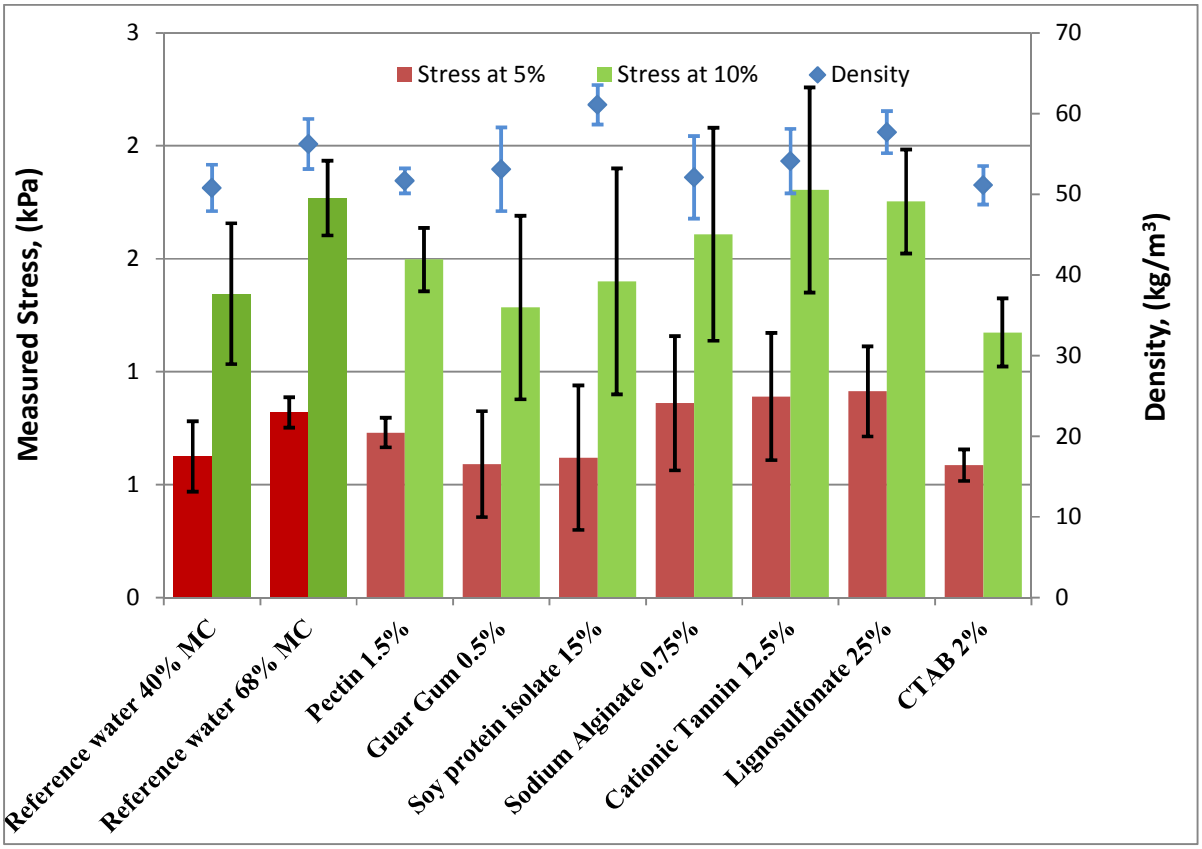


Figure 3.10: measured stress at 5% and 10% with density of treated samples.

In broad terms, the results indicate that, pectin, sodium alginate, cationic tannin, and lignosulfonate samples presented a higher mechanical resistance than the reference samples sprayed with only water at 40% dosage. Their modulus of elasticity and measured compression stresses of these samples are either similar or slightly higher than those shown for the reference 68% moisture content samples. This confirms the hypothesis that the use of binders makes it feasible to have a higher mechanical resistance than cellulose insulation sprayed with water, while using less water and, in some cases, have a slightly lower density. However, it is important to note that the high variability of measurements makes it difficult to confirm this conclusion with absolute certainty. One can see with the measured stress results that this variability is higher as strain increases. Guar gum, soy protein isolate, and CTAB treated cellulose insulation all performed slightly worse than the reference. In the case of CTAB this results was expected, as surfactants are not typically used as strength enhancing additives in cellulose. Using tensile resistance tests, Beaupré (2012) found that the addition of CTAB to paper pulp

handhseets led to decreased tensile strength. This was explained by the fact that the use of CTAB led to decreased fiber-fiber contact and hence a decreased number of fiber-fiber hydrogen bonds, which was confirmed by hydroxyl number testing.

One of the issues with projecting viscous liquids with cellulose insulation was the fact that, even though the correct liquid flow was sprayed and the same amount of water was applied, the high viscosity can make the spray pattern irregular. These irregularities can cause a heterogeneous distribution of the liquid within the fibres, making the distribution of fibres clump up and hinder its mechanical resistance. Figure 3.11 exemplifies this issue

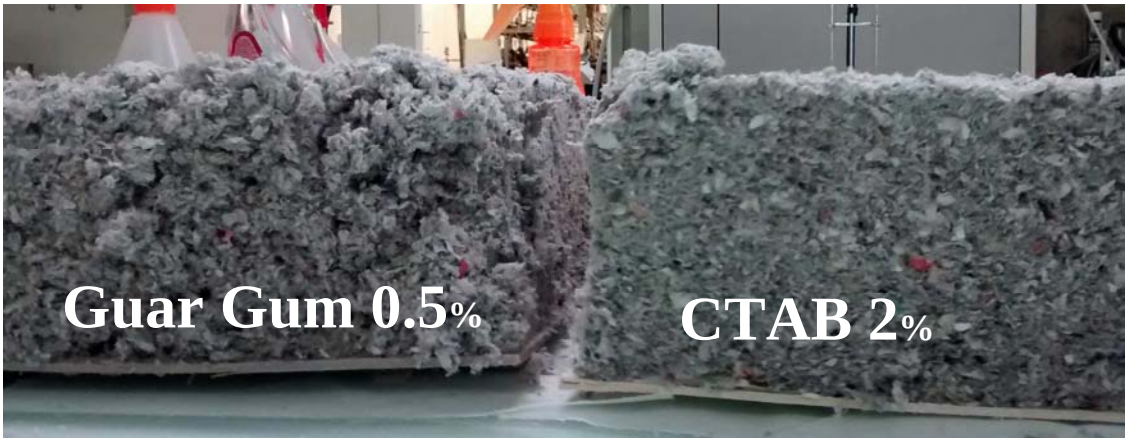


Figure 3.11: Heterogeneities of samples.

This problem was mostly prevalent with guar gum and soy protein isolate, which while showed good adhesive strength in tensile tests, ultimately became detrimental to the mechanical strength of cellulose fibre insulation due to this clumping issue. This was also present to a lesser extent with sodium alginate, which while having a high compression resistance, showed the highest standard deviation for both mechanical resistance and density. Nevertheless, despite the fact that the other two formulations (pectin and tannin) which showed improved mechanical performance could not dry faster than the reference they could be usable in future studies where drying is not a factor but mechanical performance of the material is, i.e. prefabricated cellulose insulation panels.

3.4.5 Drying of reduced moisture content samples.

Up to this point, the only additives which have been shown to improve the drying times of cellulose insulation were lignosulfonates and CTAB, with lignosulfonates having the added benefit of improving mechanical resistance. Another potential strategy to reduce drying times for cellulose insulation was to reduce the initial applied moisture, while compensating the loss of adhesive strength with an additive. The formulations which showed a higher compressive strength were sprayed at moisture content lower than the minimum of 40%. Unfortunately the final samples had problems with sagging and detachment of the cellulose. This was mainly due to the fact that since the amount of liquid was reduced, its distribution and dispersal within the material was uneven, making some parts of the material weaker and thus susceptible to separation. The low water content also made the presence of dust harder to control, reducing visibility during spraying this makes it therefore very difficult to reduce the moisture content below 40%, despite the fact that the applied additive reinforces the material. Figure 3.12 shows cellulose insulation samples sprayed with 1.5% pectin with decreasing moisture content. When moisture content falls below 39%, the material shows heterogeneous distribution of fibres and settling. The absolute minimum moisture content for cellulose insulation with just water was found to be around 38%, and 36% for cellulose sprayed with lignosulfonates, due to the increased dry solids content in the sprayed solution. As a consequence, the other candidates are adapted to wet spray cellulose. The final viable formulations remain lignosulfonate at 25% and CTAB at 2% concentrations, which will be optimized further in the following chapter.

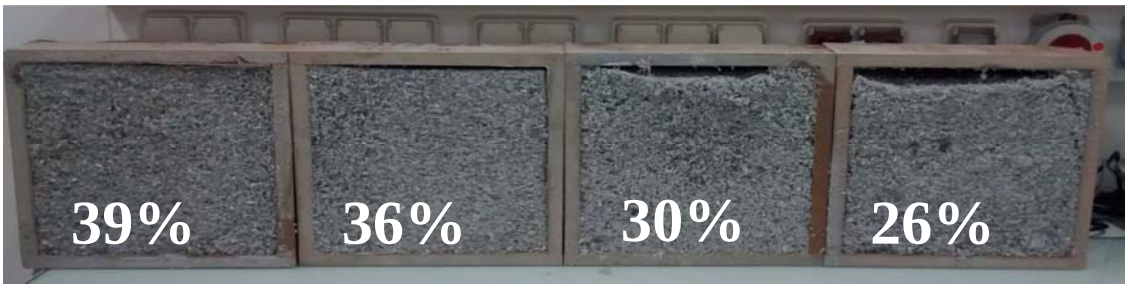


Figure 3.12: Cellulose-pectin samples sprayed at low dosage concentrations.

3.4.6 Thermal conductivity

The thermal conductivity results from chapter 2 showed the influence of initial moisture content on final thermal conductivity. An association could be found between sprayed moisture content, density, and thermal conductivity. The same analysis needs to be made with regards to the contribution of additives. Thermal conductivity at 10°C and 25°C, as well as density of sprayed samples with binders of the previously defined concentrations and 43%± 3% sprayed dosage were tested in the same method as with chapter 2 (EM12), and compared with reference samples with only water at 41% and 66% average moisture content, with 2 data points per value (Figure 3.13).

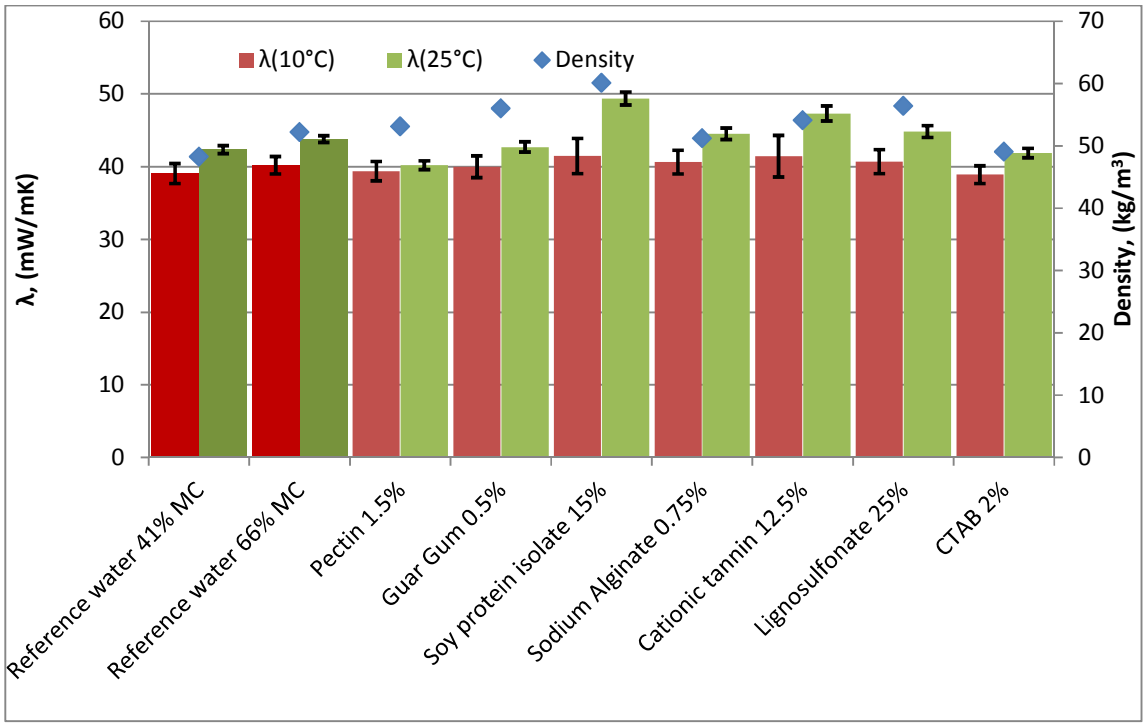


Figure 3.13: Thermal conductivity of cellulose insulation samples.

As was observed in the relation with sprayed moisture content, the increase in thermal conductivity can be linked to additive concentration and density. This relation is more defined in thermal conductivity values at 25°C, where measurements were less disperse. Pectin and guar gum, showed similar densities to the 66% moisture content samples, but lower thermal conductivity values were closer to the 41% reference samples. One reason for this could be because of the increased viscosity, the liquid only penetrates

the bigger pores of the fibres, leaving the smaller pores open. It has been shown that for materials with a similar porosity, but a lower pore size distribution, thermal conductivity will decrease (Alvarez et al., 2010). Soy protein isolate had the highest thermal conductivity and density, supporting the notion that the additive only contributes in increasing the density of the material and not reinforcing it. For the final viable formulation of lignosulfonate, an increase in the values of λ were observed, which needs to be taken into account when applying the material and studying its influence for thermal comfort. For CTAB, a slight decrease was observed, making it a better insulator, suggesting that the porosity increased with the surfactant, although the variability makes this hard to define. The formulations will be further optimized with this regard in the following chapter.

3.5 CONCLUSIONS

Building from the work from past chapters, a strategy was utilized to find a suitable additive to be incorporated with wet spray cellulose fibre insulation. First the available additives were characterized, with regards to their viscosity, and their adhesive forces. Most showed Newtonian behaviour at low concentrations, with some concentrations showing shear thinning behaviour where the sprayable viscosity threshold could be reached. Unfortunately, this discounted several binders, since viscosity is an inherent property of adhesives. Many additives which showed good adhesive properties were too viscous to be applied with the spray equipment or conversely showed a low enough viscosity to be sprayed but showed low adhesive strength. A range of non-ionic, cationic and anionic surfactants were also screened and tested with regards to their surface tension vs concentration. The additives complying with the adhesion and viscosity requirements were determined; afterwards they were tested in a bigger scale with cellulose insulation and the spraying equipment. First the candidate additives were sprayed without cellulose to ensure that the proper amount of liquid would be applied. In some cases the increase in viscosity of shear thinning liquids required a higher pump pressure to have the same flow rate. After it was verified that the additives could be sprayed at a similar flow rate than to that of water, they were applied with blown cellulose in the same conditions as done previously. They were then characterized with

regards to their drying times. With the notable exception of lignosulfonates, it was found that for binders, the increase in viscosity and the formation of a crust of the surface slowed moisture evaporation. The only case in which faster drying times was observed was through the increase of the initial dry solids content with, in particular, high concentration of lignosulfonates. Cationic surfactant CTAB was found to slightly improve these drying times as well. These samples were then tested with regards to compression modulus and thermal conductivity. Some samples were found to increase mechanical resistance at similar moisture content and density than the minimum, but unfortunately spraying binder formulations with low spraying dosage, in hopes that the increased mechanical resistance would compensate, was not feasible due to the heterogeneous distribution of moisture and a large production of dust while blowing the cellulose. Therefore, after having screened the additives with regards to their compatibility with cellulose insulation and its equipment, only lignosulfonates and CTAB remained as possible candidates. Pectin and tannin, which did not improve drying times yet showed improved mechanical performance, could be potentially used in the manufacture of prefabricated cellulose insulation batts.

Finally the feasible formulations were tested with regards to their thermal conductivity. Some additives were found to produce a denser material with lower thermal conductivity than cellulose insulation sprayed with only water with similar final density. This suggests that the inaccessibility of the smaller pores by the viscous liquid could be beneficial in the materials insulating properties. Lignosulfonates were found to increase thermal conductivity to an acceptable level, only slightly decreasing its insulating capacity. CTAB had thermal conductivity values very close to that of the reference samples.

In the following chapter we will try to further optimize the final product of cellulose insulation treated with additives. The combination of lignosulfonates and CTAB needs to be investigated. As drying tests were only done in constant controlled relative humidity, the influence of weather and insulation thickness needs to be studied for when the material is used in actual construction projects. The positive or negative influence, if any, on the material's fungal and fire resistance, as well as other practical factors, are

important issues to consider moving forward when characterizing the final insulating material.

4

FINAL PROPERTIES OF INSULATING MATERIAL AND HYGROTHERMAL MODELLING

4.1 INTRODUCTION

Formulations of wet spray cellulose insulation have been established, in which additives improved the drying and mechanical performance of the material. Both lignosulfonate and CTAB have been found to reduce drying times, while either improving or maintaining the mechanical resistance of cellulose insulation. However, the formulation could still be optimized, in which a compromise could be made between the desired decrease in drying times and the amount of additive used. The influence of the concentration of lignosulfonate and CTAB additives needs to be established to reduce cost of the final product and its impact on density and thermal conductivity.

The previous work has established the drying performance of cellulose insulation samples with 10cm thickness and at constant, controlled relative humidity conditions. A more comprehensive study needs to be done in which the influence of thickness and weather conditions need to be taken into account in order to fully understand the impact of the included additive under real conditions, before it can be applied at a construction site. Specific numerical tools and models have been designed for this purpose, in which these factors could be assessed.

Thermal insulation materials such as cellulose are subject to numerous national and international standards. Some of these standards specify the required fungal and fire resistance requirements for a material to be used as an insulation material. It is therefore important to confirm that the included additives do not hinder these properties and if possible, improve them. This could make the additive a partial or complete replacement of the mineral additives already used, mainly borate salts.

In this chapter, the influence of the concentration of additive candidates on the drying, mechanical, and thermal properties of the final product are studied. The influence of the thickness and ambient conditions on drying of the final formulation is studied using hygrothermal modelling software. Finally, preliminary studies on the material's fire performance were done to indicate the influence of the additives in this regard.

4.2 Influence of additive dosage on the properties of cellulose insulation

Both additives have a low viscosity and good solubility in water, making it feasible to mix them with cellulose at high concentration. It is important to determine the optimal concentrations. Formulations of cellulose insulation with lignosulfonate and at different concentrations were studied with regards to drying, compression resistance, and thermal conductivity. Concentrations of lignosulfonate ranged from 10% to 40%, and for CTAB from 1% to 4%, for which higher concentrations could not be reached due to high foam production.

4.2.1 Drying

The drying of sprayed samples as a function of lignosulfonate and CTAB concentration is shown in in Figure 4.1 (only CTAB at 2% is shown since the other concentrations were indistinguishable from the reference). The interpolated time to reach a dry state is shown in Table 4.1.

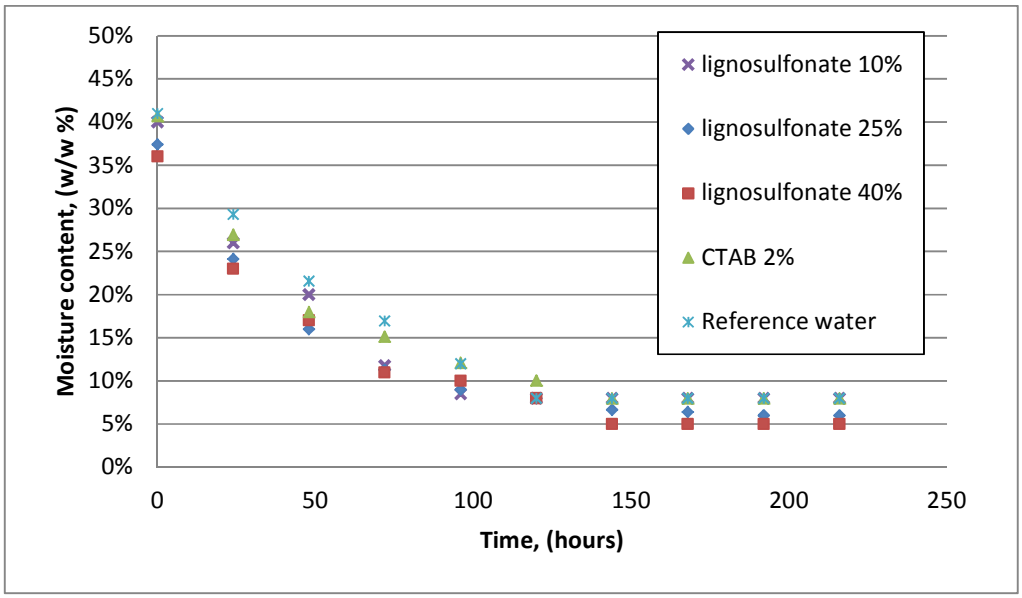


Figure 4.1: Drying cure of cellulose with lignosulfonate additives at variable concentration.

Formulation	Time to reach 20% MC, (hours)
Reference	51
Lignosulfonate 40%	34
Lignosulfonate 25%	38
Lignosulfonate 10%	49
CTAB 1%	47
CTAB 2%	45
CTAB 4%	48

Table 4.1: Time to reach dry state for cellulose with lignosulfonate additives at variable concentration.

The initial and equilibrium moisture decreases with concentration of lignosulfonate, which is indicative of the increased dry solids content of the material. This factor is the main impact parameter to the reduction of drying times. The influence of 10% concentration of lignosulfonates would seem to be negligible with regards to drying. In a practical sense, the minimum effective reduction in drying times would be 15% which roughly translates to 1 day per week of drying. Assuming a linear relation in drying times between 10 and 25% moisture content, a reduction of the reference drying time by 15% (42.5 hours of drying to reach 20%), would require a concentration of 19%. For the surfactant formulations, higher concentrations of CTAB were less impactful,

leaving 2% as the optimum. Considering the fact that both additives use different mechanisms to improve the drying performance of cellulose insulation, it would be feasible that a combination of both additives could improve drying times even further. The mix of additives, CTAB and lignosulfonate, was also tested, but results showed no difference between the drying with lignosulfonate and lignosulfonate mixed with CTAB. This could be due to the circumstance that lignosulfonate also has surfactant properties. Surface tension measurements (EM16) of just lignosulfonate at 25% concentration in water found a surface tension value of 56 mN.m.

4.2.2 Compression resistance

Compression tests of samples with different lignosulfonate and CTAB concentrations were tested according to the method used in previous chapters (EM11). The elastic modulus of tested samples is shown in Figure 4.2. All lignosulfonate samples showed improved mechanical resistance; therefore even though the low concentration samples showed little reduction in drying times, they ensure an improved resistance to settling.

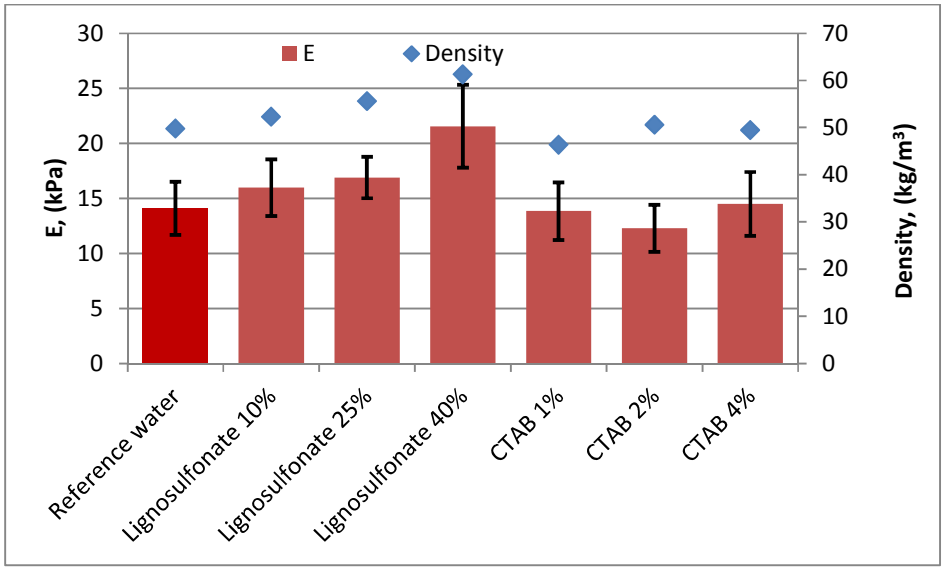


Figure 4.2: Compression resistance for cellulose with lignosulfonate and CTAB additives at variable concentrations.

An issue occurred with the 40% samples where the high concentration led to heterogeneous distribution of the binder, which explains the high standard deviation in

values. The range of 19-25% concentration for lignosulfonate seems to be adequate. As expected, CTAB samples showed no improvement in compression resistance.

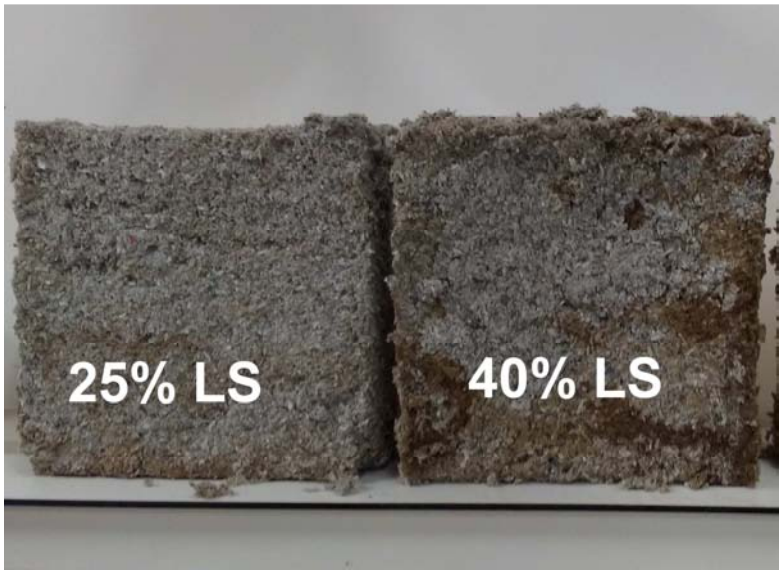


Figure 4.3: Heterogeneity in 40% concentration lignosulfonate samples

4.2.3 Thermal conductivity

Thermal conductivity measurements on samples with varying lignosulfonate and concentration were also tested, using the method from EM12. Results for thermal conductivity and density as a function of CTAB and lignosulfonate concentration are shown in Figure 4.3. The addition of lignosulfonate increased thermal conductivities with increasing concentration, again both due to the increase in density and the closure of smaller pores within the fibre. CTAB formulations had similar thermal conductivity values to the reference, with slightly lower values at 4% concentration, although the variations are significative compared to the reference, making the lower values are within the margin of error.

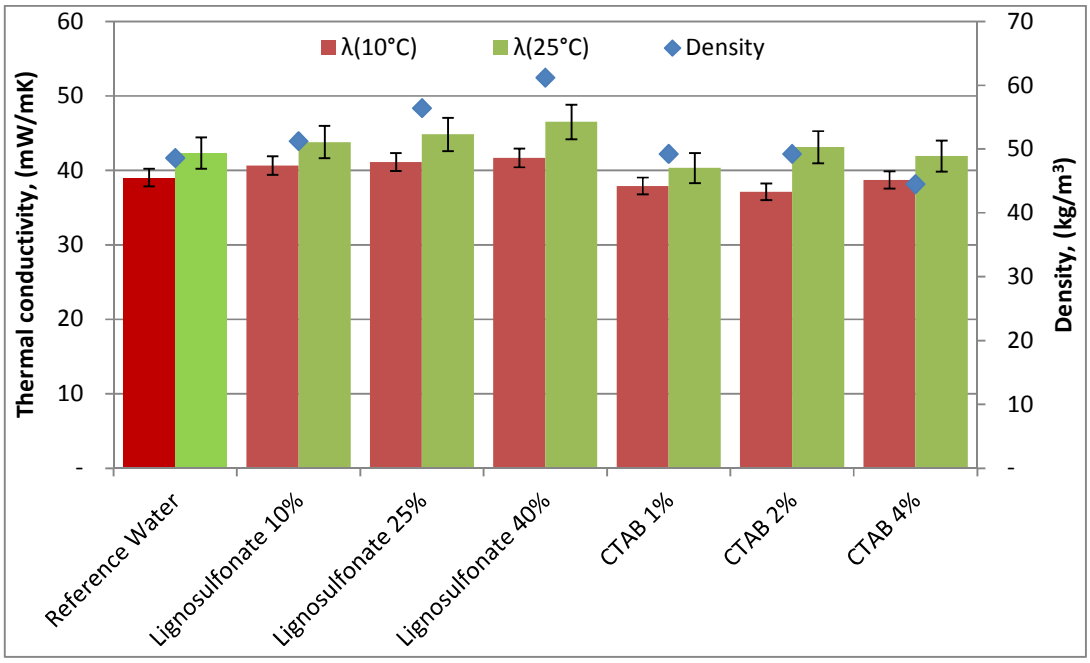


Figure 4.4: Thermal conductivity for cellulose with lignosulfonate and CTAB additives at variable concentrations.

4.3 CALCULATED DRYING PERFORMANCE OF TREATED AND UNTREATED CFI

Up until now, there has been no fixed method or standard to determine the time required for wet spray cellulose insulation to reach a dry state. In the French technical evaluation for a cellulose insulation material, there are indicative assessments on the drying times for cellulose, as a function of thickness and season it is applied (CSTB, 2011).

Time to reach dry state, (days)			
Thickness, (mm)	Season of installation		
	Summer	Spring/Autumn	Winter
60	3	6	8
90	3	7	9
120	4	8	11
150	5	10	13
180	7	12	15
200	9	14	17

Table 4.2: Indicative drying times for cellulose insulation as a function of thickness and ambient conditions (CSTB, 2011)

While these are good indicators of drying times, they are not very accurate and do not take into account the initial sprayed moisture, which can vary depending on the applier. Also for the new formulations the impact of the additive on drying times for all of these cases (initial moisture, thickness, ambient conditions, etc.) needs to be considered.

Usually, a builder periodically measures the moisture content on the material using a portable moisture meter. Ideally, a cellulose insulation applier should be able to predict drying times for the material reliably, without having to constantly measure moisture content or rely on drying times which could be overestimated. There exist several models which could be used in this regard, where the conditions in which the material is installed could be taken into account. First a simple empirical model will be used to evaluate each parameter individually and its influence on drying under static conditions. Then numerical modelling software will be used to determine drying times using material parameters, dynamic temperature and humidity conditions. The hygrothermal performance of the material once dry will also be studied.

4.3.1 Empirical model

Several empirical and semi-empirical mathematical models to characterize the drying of building materials over time have been proposed. These models and their effectiveness for certain types of building materials were reviewed by Barreira et al. (2014). A simple model is proposed by Delgado et al. (2006), using a isothermal first order equation:

$$\frac{dw}{dt} = \frac{1}{t_c}\left(w_{eq} - w(t)\right)$$

(4.1)

Where: $\frac{dw}{dt}$ is the drying rate, w(t) is the material moisture content at time t, t_c is known as the drying time constant, and w_{eq} is the moisture content at equilibrium for the material. Using static drying conditions, integrating this equation for the initial and end conditions t = 0 to t = t and w = w_0 to w = w, where w_0 is the initial moisture content, gives:

$$w = w_{eq} + \left(w_0 - w_{eq}\right).\exp\left(-\frac{t}{t_c}\right)$$

(4.2)

Applying this equation to the drying of cellulose insulation, it is possible to determine an empirical equation for the moisture content of the material w, as a function of time. The previous chapters have shown that the equilibrium moisture content for wet sprayed cellulose insulation w_{eq}, tends to be around 8% regardless of initial sprayed dosage, w_0. The only unknown parameter in equation (4.2) is the drying time constant t_c, which is sometimes attributed to the time required to dry 2/3 of initial moisture, could be determined by curve fitting experimental data from previous chapters with the empirical model.

4.3.1.1 Influence of sprayed moisture.

The results from 2.8.2 showed the influence of installed moisture content on drying. The values from these measurements were fitted with the first order empirical model from equation 3.2. The drying constant t_c was estimated using the root mean square error (RMSE) between the experimental and the predicted values using the solver function with excel (See EM17 for more information). The curves were then plotted against the measured values, as shown in Figure 4.4.

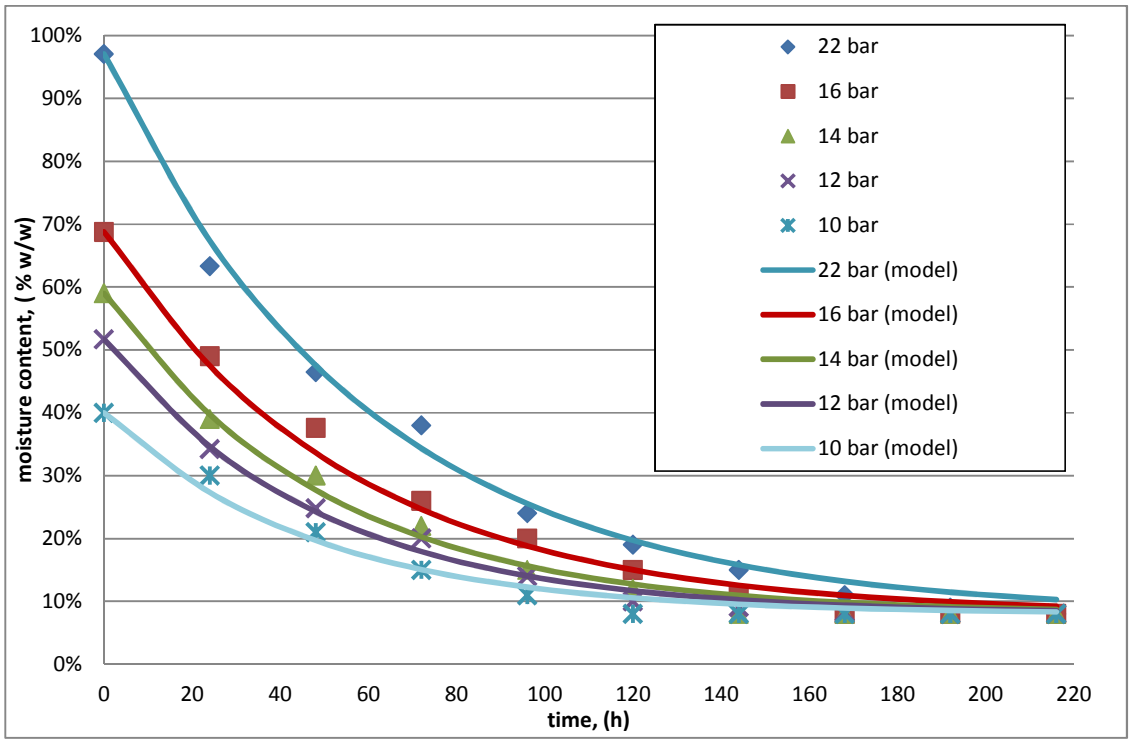


Figure 4.4: Plotted drying times with equivalent empirical model of cellulose with varying initial dosage.

The model shows good correlation with measured values. Table 4.3 shows the values of initial moisture content w_0 as well as the fitted values of t_c with the RMSE (n=10) for each curve, with Figure 4.5 showing the relation between both constants. The more the initial moisture content, the higher the error, indicating less reliability of the model at high initial dosages.

Applied Water pressure	Initial moisture w_0	w_{eq}	t_c	RMSE
22 bar	97%	0.08	59.14	0.0052
16 bar	69%	0.08	55.65	0.00403
14 bar	59%	0.08	50.58	0.002352
12 bar	52%	0.08	48.64	0.00111
10 bar	40%	0.08	47.71	0.002083

Table 4.3: Constants for empirical model for drying of cellulose with initial moisture dosage.

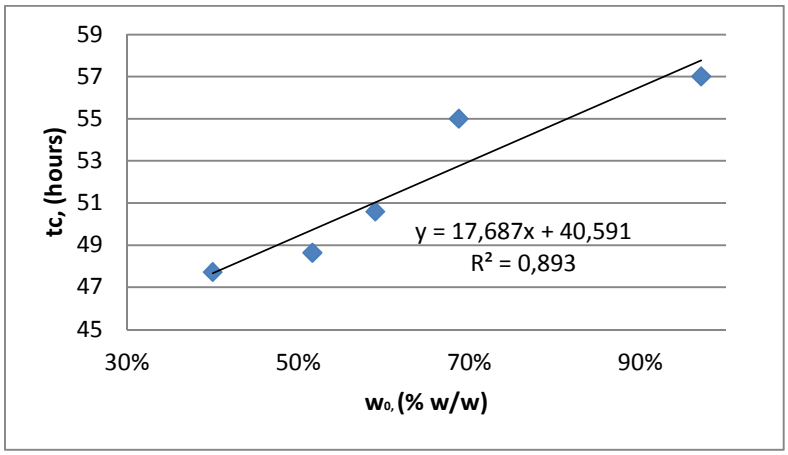


Figure 4.5: Influence of initial moisture on the drying time constant $t_{c.}$

The value of t_c increases with increasing sprayed moisture content. With this into account it is possible to consider the relation between both values as linear, thus making it possible to consider the drying curve as a function of only the initial and equilibrium moisture contents with the interpolation of $t_c=f(w_0)$. Equation 4.3 then becomes:

$$w = w_{eq} + (w_0 - w_{eq}).\exp\left(-\frac{t}{17.687w_0 + 40.591}\right) \quad (4.3)$$

With time t in hours and w_0 in (% w/w). This could be useful in characterising the drying performance of cellulose insulation as a function of installed moisture. A cellulose insulation applier could estimate the time required to dry the material from just an initial moisture reading. Ideally the installed moisture content for cellulose insulation should always be the minimal 40% because the influence of additive at high moisture would be negligible when compared to cellulose sprayed with just water at the minimum of 40%. In other words, cellulose without additive at 40% moisture constant will always dry faster than cellulose with lignosulphonate installed at 60% moisture content.

4.3.1.2 Influence of insulation thickness

When used in a building envelope, the installed thickness of an insulation material depends on the required thermal resistance for a building's interior. The thickness of cellulose insulation has an effect on the drying times because the moisture has to go through more of the porous material to evaporate to the surface. Standard sprayed cellulose insulation samples at around 40% ± 3% moisture content and of 100, 150, and

200 mm thickness were dried according to the method used in EM10, with samples weighed periodically. The drying curves were again fitted with the empirical model for specimens of different thicknesses. Figure 4.6 show the drying curves with the fitted empirical models.

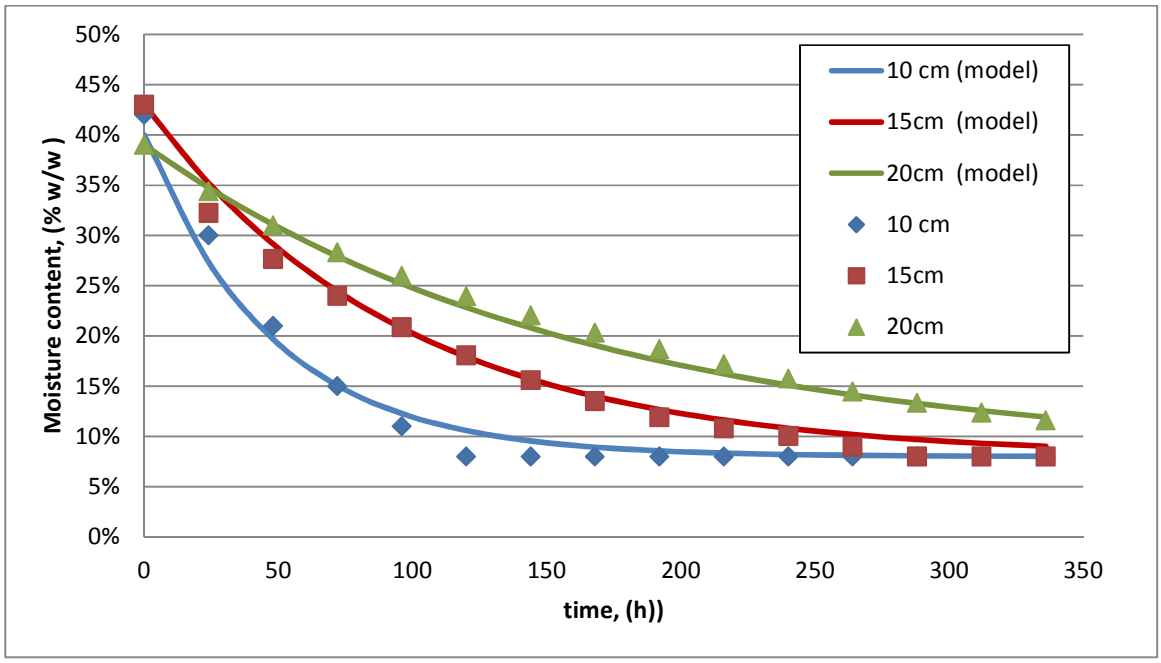


Figure 4.6: Plotted drying times with equivalent empirical model of cellulose with varying thickness.

Again, a good correlation is found between measured results and predicted values. Figure 4.7 shows the plot of sample thickness vs the estimated values of t_c. It is then possible to adapt equation 3.1 to estimate the drying times of samples sprayed at the minimal moisture content 40 % as a function of sample thickness, e (in cm).

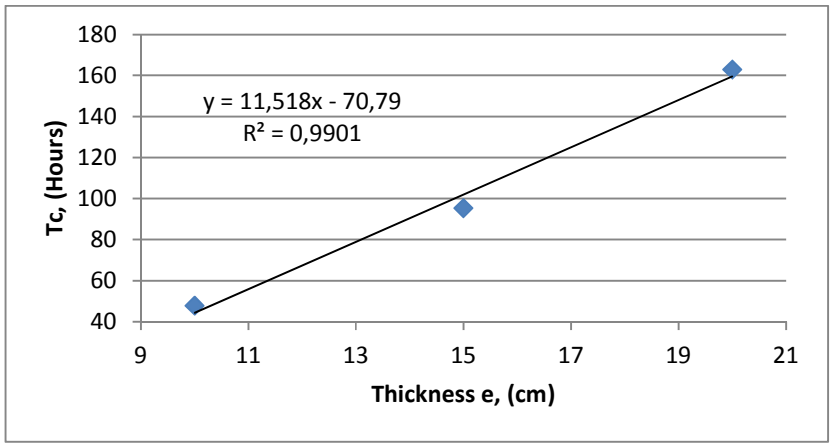


Figure 4.7: Influence of installed thickness on the drying time constant, t_c

$$w = w_{eq} + (w_0 - w_{eq}).\exp\left(-\frac{t}{11.518e - 70.79}\right)$$

(4.4)

The relative influence of the thickness of the insulation sample on drying times is higher than that of installed moisture. However, unlike the installed moisture content, the cellulose thickness is imposed by the requirements in insulation performance for a building, and not a result of improper application of the material.

4.3.1.3 Influence of additives.

The drying curves from section 4.2.1 were fitted with the empirical formula. The differences in initial and equilibrium moisture content for each formulation were taken into account. Table 4.4 shows the value of the drying constant for each formulation, with the initial and equilibrium moisture contents.

	Initial moisture (%w/w)	Moisture at equilibrium (%w/w)	Tc (hours)
lignosulfonate 10%	40	0.08	39.09
lignosulfonate 25%	37	0.06	41.75
lignosulfonate 40%	36	0.05	45.73
CTAB 2%	41	0.08	43.21
Reference water	41	0.08	47.71

Table 4.4: Constants for empirical model for additive formulations.

The drying time constants for all formulations were found to be lower than that of the reference sample. Interestingly, the drying time constant increased with increasing

lignosulfonate content, indicating that the use of the additive actually reduces the slope of the drying curve, but the reduction in initial moisture content allows faster drying. When comparing with the time constants from moisture dosage and cellulose thickness, it is evident that these initial conditions have much more of an effect on the drying of cellulose insulation than the use of addittives.

Considering that the first order model showed drying curves very close to experimental results, this approach could be a way to evaluate drying times as a function of both installed moisture content and thickness, which would allow optimized construction schedules since no extra time would be spent waiting for the material to dry. The same logic could be used when studying the influence of other factors such as temperature, humidity, and orientation of the material. In order to take these factors into account a more comprehensive model needs to be used, which can take into account not only the material's properties, but also the atmospheric conditions when it is being applied.

4.3.2 Numerical model: WUFI software.

While the empirical model has shown to provide a good indication of drying times, it relies on experimental results under static conditions to model the drying behaviour of cellulose. In order to take into account other variables such as temperature and humidity, a more comprehensive model is necessary. WUFI 5, developed by Fraunhofer Institute for Building Physics (Stuttgart, Germany), is a hygrothermal numerical modelling software which could be used in this regard. It uses a comprehensive coupled heat and mass transport model which has been validated by Kehrer and Schmidt (2008) and others. Using the material properties and different ambient humidity and temperature scenarios, the hygrothermal performance, during and after installation of cellulose insulation, could be evaluated.

4.3.2.1 Theory

WUFI 5 software uses a finite elements method to solve the coupled heat and mass transport equations (Karagiozis et al., 2001):

Energy transfer

$$\frac{\partial H}{\partial T} \cdot \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + h_v \nabla \cdot (\delta_p \nabla (\phi p_{sat}))$$

(4.5)

Moisture transfer

$$\frac{\partial w}{\partial \phi} \cdot \frac{\partial \phi}{\partial t} = \nabla \cdot (D_\phi \nabla \phi + \delta_p \nabla (\phi p_{sat}))$$

(4.6)

Where: H is the total enthalpy, T is temperature, k is the thermal conductivity, h_v is the latent heat if phase change, δ_p is the vapour permeability, φ the relative humidity, w the moisture content, D_φ is the moisture diffusion coefficient. Using these equations, coupled with the initial and boundary conditions WUFI 5 can determine the heat and moisture fluxes, as well as the transient heat, relative humidity and moisture content profiles for a material or series of materials in one dimension.

4.3.2.2 Input parameters

In order to determine the hygrothermal drying performance of cellulose insulation using WUFI, the material input parameters of the model must be defined. Most of these parameters have either been determined previously, or can be taken from previous works, while others require further research. The temperature dependant thermal conductivity and density at equilibrium for both cellulose with and without additive were detailed in sections 2.2.11 and 4.2.3 respectively. While it's not essential for calculations, the software also asks for moisture dependant thermal conductivity. Unfortunately, high moisture samples could not be tested reliably using the standard equipment, so a linear relation was used based on the results found by Sandberg (1992), where:

$$\lambda_{moist} = \lambda_{dry} + 0.0002' w \text{ (W/mK)}$$

(4.7)

Where w is the mass of water per unit volume of cellulose (kg/m^3), and λ_{dry} is determined using the results from 25°C thermal conductivity measurements, taking into account the equilibrium moisture content during measurement. The porosity of the material was considered to be 0.95, as has been referenced by Binder et al. (2007). The

material moisture transport function has been determined from isothermal dynamic vapour sorption measurements from section 2.15, the software does not distinguish between sorption and desorption, and requires values close to saturation, so the Oswin model will be used, up to values of 0.9999 moisture activity. The water vapour resistance factor used was 1.8, as given by (Cerolini et al., 2009). The remaining properties which need to be determined are the specific heat capacity and the liquid transport coefficients.

4.3.2.3 Specific heat capacity

The specific heat capacity c_p is relative to heat the energy required for a material to raise its temperature by one degree Kelvin. For building materials it contributes to the reduction of peak heating loads in the summer. This is quantified by the thermal effusivity of an insulated wall. The specific heat capacity at 25°C of loose cellulose fibres was done using differential scanning calorimetry, as detailed in EM18. Values for treated and untreated cellulose insulation are shown in Table 4.5. It was shown that the addition of lignosulfonates had a slight contribution to the specific heat capacity of cellulose. No difference between CTAB and the reference samples

Sample	cp (J/kg°C) at 25°C
Sprayed Cellulose (reference water)	2.145
Sprayed Cellulose with lignosulfonate (25% concentration)	2.256
Sprayed Cellulose with CTAB (2% concentration)	2.145

Table 4.5: Specific heat capacity of cellulose insulation formulations.

4.3.2.4 Liquid transport coefficients.

The liquid transport coefficients are the most important parameters when defining the diffusion of water within a porous material. The diffusivity or diffusion coefficient D is characterized in Fick's first law where it defines the relation between the moisture flux J from high to low concentration to the moisture gradient: $\frac{\partial \varphi}{\partial x}$

$$J = -D\frac{\partial \varphi}{\partial x}$$

(4.8)

It is measured in meters per second squared. While for some applications, using a constant or reference diffusivity value is sufficient, for hygroscopic porous materials such as cellulose, moisture diffusivity is highly dependent on moisture content. Values of moisture dependent diffusivity for cellulose insulation has been determined by Marchand and Kumaran (1994) using gamma ray attenuation. Other methods for characterising diffusivity in porous materials include Nuclear magnetic resonance (NMR) spectroscopy (Pel et al., 1996). A much simpler method to approximate the values of moisture dependent diffusivity coefficients has been used by Künzel (1995). The method uses capillary sorption measurements to determine the moisture sorption coefficient A_w, measured in m^2/s, and designates the following relation between the moisture sorption coefficient, the moisture content and the diffusivity D:

$$D_l^w = 3.8\left(\frac{A_w}{w_{cap}}\right)^2 1{,}000^{\frac{w}{w_{cap}}-1}$$

(4.9)

Where w is the moisture content and w_{capt} is the moisture content at saturation of the material.The coefficient for loose cellulose insulation A_w was determined according to capillary transport method defined by the standard EN ISO 15148, (see EM19 for more details). The density of the cellulose before sorption was 52 kg/m^3. The value of the capillary sorption coefficient A_w corresponds to the first slope the moisture flux Q, (kg/m^2) during the fast period of capillary moisture sorption tests, plotted with the square root of time $t^{1/2}$ (Figure 4.8).

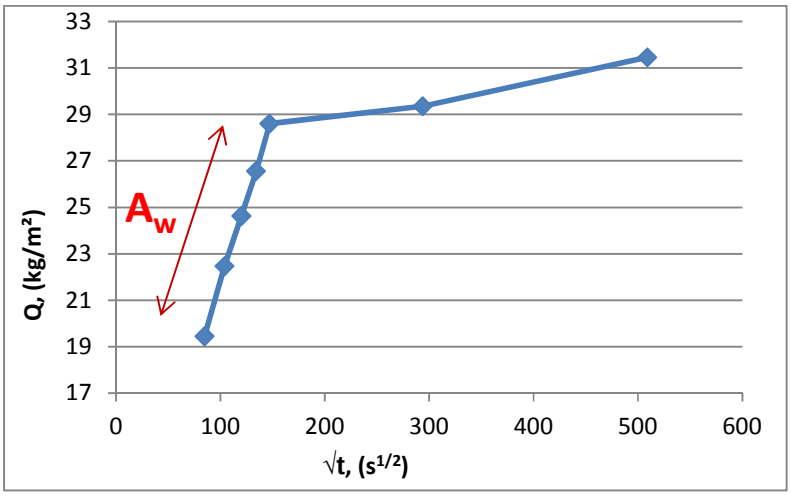


Figure 4.8: Capillary sorption measurement with sorption coefficient A_w.

The sorption coefficient A_w was found to be 0.915 kg/m²s$^{1/2}$. This value is higher than was found by Hansen et al. (Hansen et al., 2001) (0.81 kg/m²s$^{1/2}$), but a different type of cellulose was tested. The moisture dependent diffusivity coefficient can then be plotted as a function of moisture relative to saturation moisture, using equation 4.9. The value for saturated moisture content was taken from the Oswin model of isotherms (Figure 2.6), giving a value 742% w/w of moisture content at a relative humidity close to 100% (saturation). Figure 4.9 shows the calculated moisture diffusivity as a function of moisture content for loose cellulose insulation.

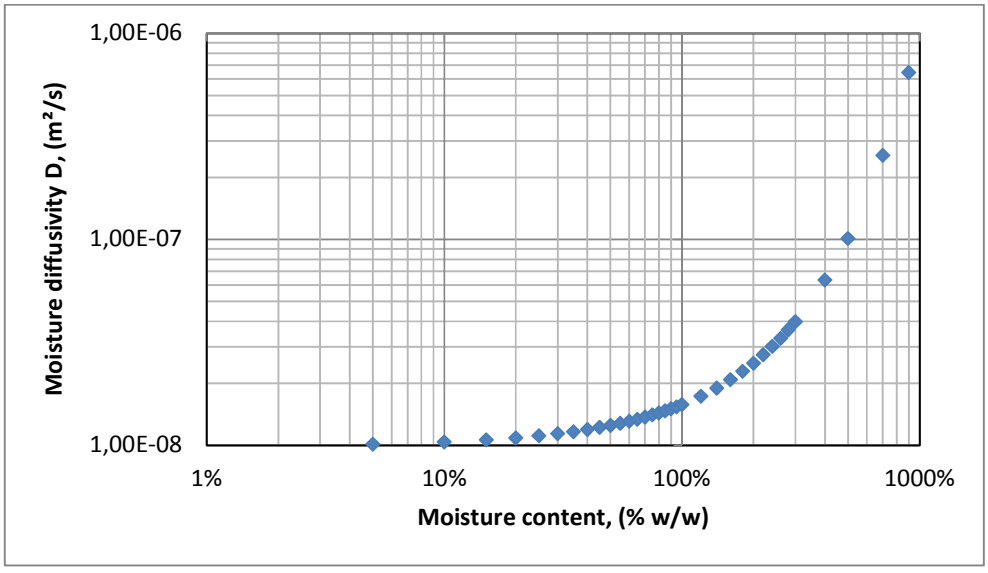


Figure 4.9: Moisture dependent moisture diffusivity coefficient for cellulose insulation.

As can be seen, diffusivity increases exponentially with increasing moisture content, which is logical since moisture diffuses faster with the higher capillary pressure. The orders of magnitude seem similar to those found by Marchand and Kumaran, (1994), as well as the reference values for cellulose insulation given in the WUFI software. The input parameters from the WUFI software distinguish sorption diffusivity D_s, for when the moisture is first absorbed within the material, and redistribution diffusivity D_r for when moisture disperses within the material. For cellulose the redistribution coefficients only becomes apparent when close to saturation, so for the drying of cellulose after it is spread, the redistribution diffusion coefficient is the same as the sorption diffusion.

The same approach for calculating the diffusion coefficients was applied for the formulations with lignosulfonate, i.e. the sorbed water in the capillary sorption tests was replaced with 25% lignosulfonate and 2% CTAB (Table 4.6). For lignosulfonate, a lower sorption coefficient was due to the higher viscosity and the fact that the moisture diffusivity during sorption is different than the one during drying since the lignosulfonate portion of the absorbed liquid stays within the fibres during drying. In the case of CTAB the sorption results yielded values slightly higher to those with just water which translates to higher diffusivity i.e. faster drying. This method is useful as a first approach in determining moisture dependant diffusivity values. The diffusivity function then needs to be adjusted according to the experimental drying curves.

Sorbed liquid	A_w $kg/m^2s^{1/2}$
Water	0.91
Lignosulfonate 25%	0.68
CTAB 2%	0.94

Table 4.6: Measured capillary sorption coefficient $A_{w.}$

4.3.2.5 Validation of parameters

In order to determine the accuracy of the models, most notably the diffusivity D, a simulation was made using the controlled experimental conditions (25°C, 60%RH).

Using the material parameters determined previously, models of the drying of cellulose samples were established and compared to experimental results. The drying of a 100mm cellulose sample with initial 40% moisture content. In order to have the drying of the material be unilateral, a 1mm layer of vapour barrier membrane was added to the left of the cellulose material. The duration of the model was 10 days. The drying curve from the WUFI model using the estimated liquid transport coefficients from capillary sorption tests was compared to the measured experimental results. Using equation 4.7, the value of the moisture sorption coefficient was adjusted in a way to provide a moisture diffusivity function in which the difference between measured and calculated moisture values was minimal. This approach was used previously, by Krus and Holm (1999). Figure 4.10 shows the calculated drying curves of cellulose insulation at 40% initial moisture, compared with the experimental results determined previously.

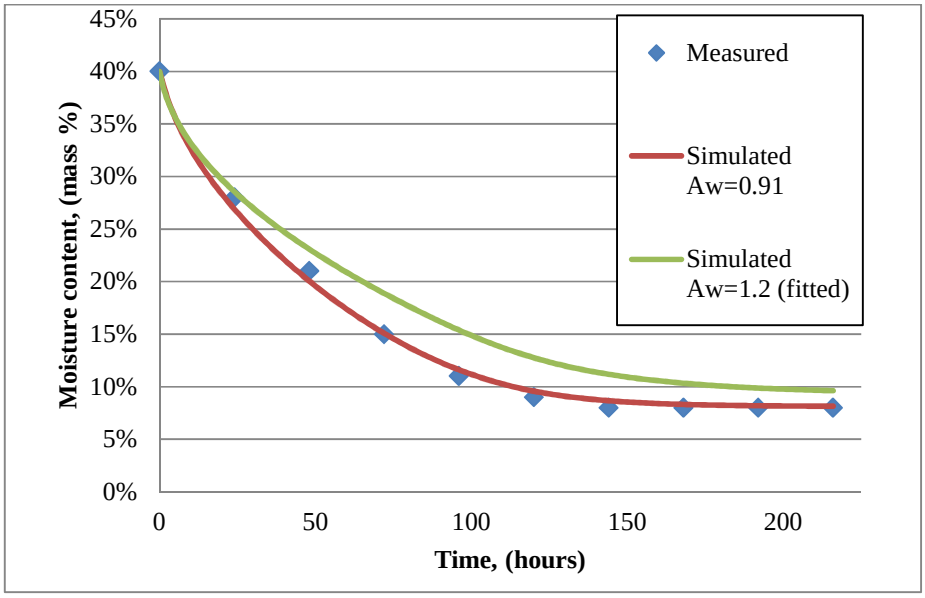


Figure 4.10: Drying curves of cellulose with calculated and fitted capillary sorption constant $A_{w..}$

The same approach was used to fit a moisture diffusivity function that would be representative of the drying of cellulose with included additives. Table 4.7 shows the new fitted values of A_w for the formulations of cellulose insulation. Both had slightly higher values indicating an increase in moisture transport, though the lignosulfonate formulation has the added benefit of requiring less initial moisture.

Formulation	A_w (fitted) $kg/m^2s^{1/2}$
Water	1.15
Lignosulfonate 25%	1.3
CTAB 2%	1.35

Table 4.7: Values of fitted capillary sorption constant A_w

4.3.3 Drying performance of CFI under different installation conditions

Having established and validated the required characteristics for the drying model in WUFI, a more in depth study can be made on its drying performance with real world climate conditions. In order to quantify the influence of the additive in the reduction of drying times for different weather and insulation thickness conditions, a series of 24 simulations with varying parameters were made, comparing cellulose sprayed at the 40% moisture content with either the reference water or the 25% lignosulfonate additive (LS 25%), varying insulation thickness (10 cm, and 20 cm), initial moisture dosage (40% "normal dosage or 80% "high dosage") and weather profiles (summer, autumn and winter). Only the lignosulfonate formulation was tested to measure the highest feasible reduction in drying times.

The following material profile was used, from exterior to interior (Figure 4.11): a 1mm weather barrier membrane, a 15mm oriented strand board (OSB) panel (density: 595 kg/m^3) from which the properties are from the internal software material database, and finally the cellulose insulation of varying thickness (see EM 20).

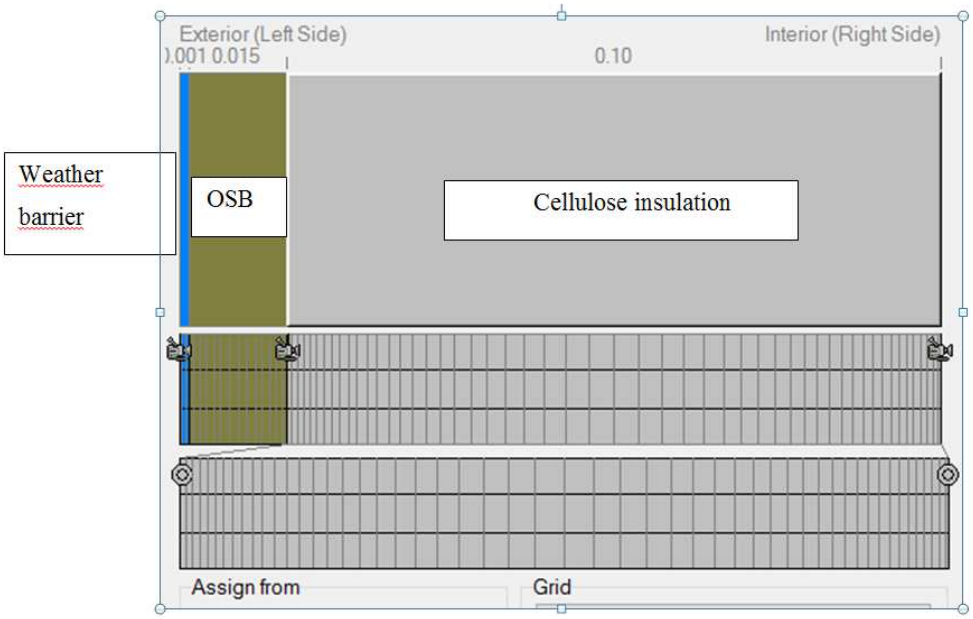


Figure 4.11: WUFI wall material layers profile.

In order to input the weather profiles, WUFI provides climate data for various cities around Europe, Asia, and U.S. For this study the climate data from the city of Bordeaux, France, the city in which the studied cellulose material (Univercell Comfort) is produced, was used. The climate data from the months of July, October, and January 2008 were used for the weather profiles of summer autumn and winter, respectively. The average moisture and temperature conditions for each period were 67% RH, 19°C in summer, 74% RH, 14°C in autumn, and 84% RH 5°C in winter. These weather conditions were applied on both sides of the cellulose wall. The wall was considered to be orientated south. All simulations had a duration of 30 days, unless the threshold of 20% moisture was not reached, in which they were extended to 60 days.

Figures 4.12 and 4.13 show the simulated drying curves of cellulose insulation with varying parameters, (sprayed liquid, thickness, and period of application) for initial sprayed dosages of 40%, and 80%, respectively. The drying times vary greatly according to these factors, ranging from the quickest: cellulose with lignosulfonate sprayed during summer at 10cm thickness, to the slowest: cellulose with water sprayed during winter at 80% moisture content and 20 cm thickness. The ambient conditions had a high impact on the equilibrium moisture content of the material, some samples took longer than the standard 30 days of simulation to reach the 20% moisture threshold. The contribution of

the additive to the reduction in drying times for each situation is apparent, but can vary greatly depending on the parameters. When associating with the drying curves obtained from the empirical model in the previous section, a relationship between the empirical model and drying times at summer conditions can be found, confirming the notion that the model provides a good initial approach in predicting drying times.

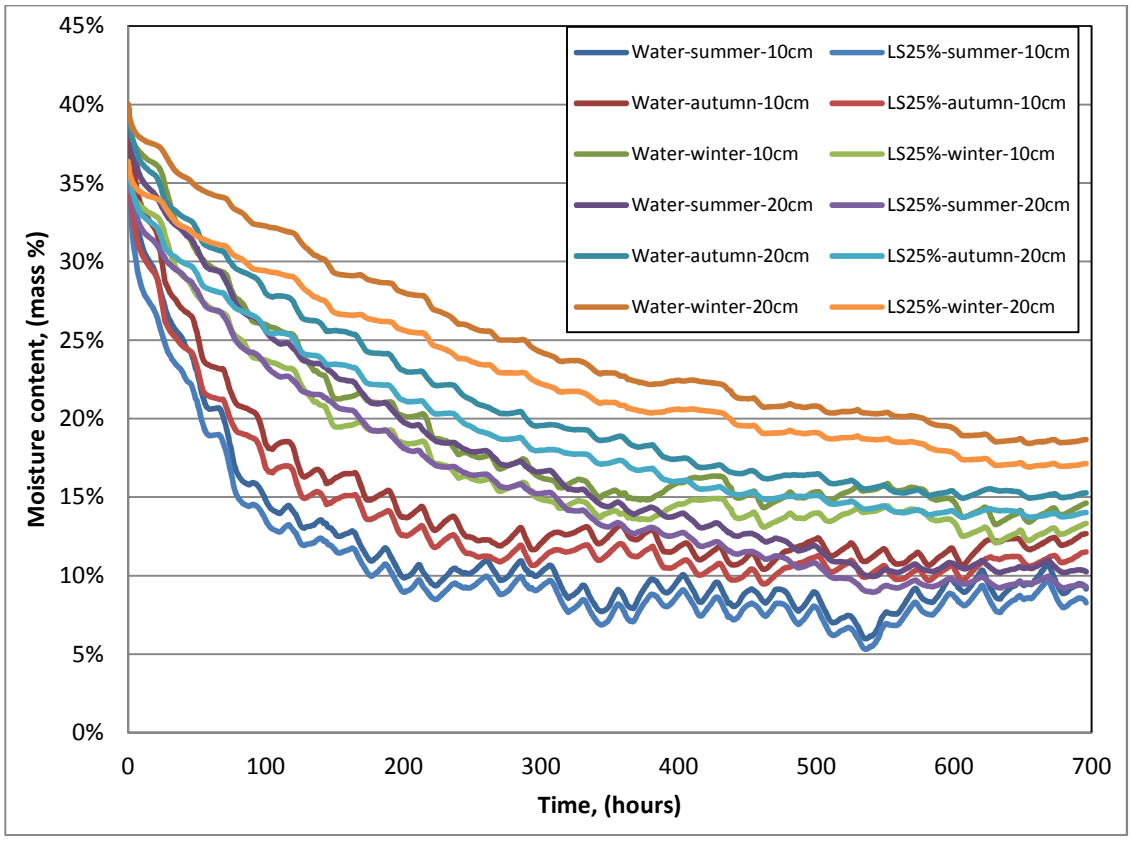


Figure 4.12: Modelled drying curves under different ambient and thickness conditions at an initial dosage of 40%

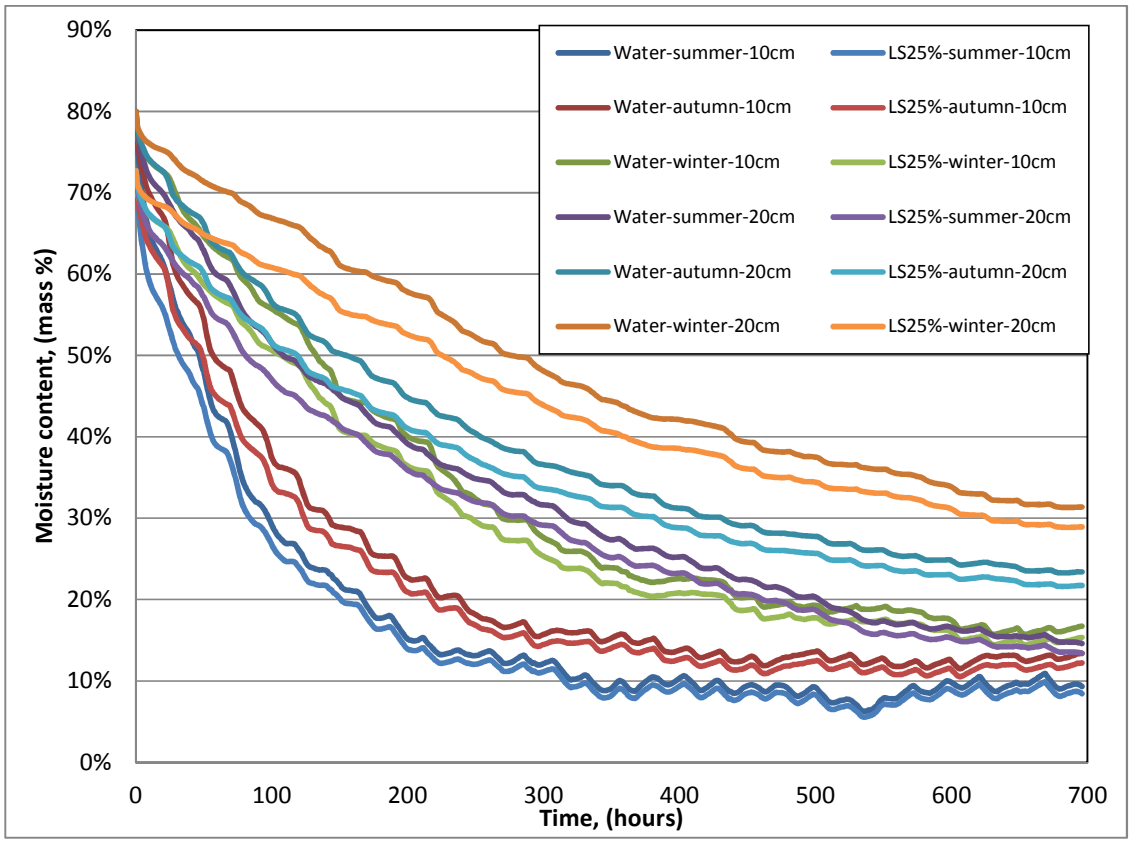


Figure 4.13: Modelled drying curves under different ambient and thickness conditions at an initial dosage of 80%

Figure 4.14 shows the time to reach 20% moisture content, measured in days, for all simulations. The relative decrease in these times for lignosulfonate treated walls, compared to the reference cellulose with water is shown in Table 4.8. A steep increase in time required to reach the dry state can be observed when cellulose is projected at 80% dosage and 20 cm thickness. These cases require between twice and four times the required drying times of 20 cm cellulose sprayed at the proper 40% dosage. The appropriate application of material is therefore crucial at higher insulation thicknesses. The effectiveness of the additive is also reduced at higher dosages, with a reduction in drying times of only 5-10% for samples sprayed at 80% dosage. The condition in which the additive was most efficient in the reduction of drying times was during winter using the minimal sprayed dosage of 40%. While the relative improvement in drying times for 10 cm cellulose sprayed with lignosulfonate additive at 40% in summer and autumn is high, this translates to a reduction in drying times of just 16 and 20 hours respectively. In

practical terms this reduction might still not be negligible, since the cellulose would dry in the morning instead of the evening, therefore effectively saving one work day in the construction site for the installation of a vapour barrier. Moreover, the improvement in mechanical resistance is also an added benefit. Nevertheless this improvement might not be cost effective. Suggesting that the additive should improve drying times by at least 24 hours despite weather conditions, and interpolating between reductions in drying times from 10 to 20 cm, it's possible to deduce that the minimum insulation thickness in which lignosulfonate additive would be effective at 40% sprayed liquid dosage would be 16 cm in summer conditions and 12 cm during autumn conditions, below that thickness, it could still be used, but its main purpose would be to strengthen the material at a lower moisture content.

The drying performance of cellulose insulation needs to be established by means of full scale tests under real conditions and compared with the models. Ideally, using ambient temperature and humidity readings, coupled with moisture meters within the material, the model could be validated with on-site results. Unfortunately due to time constraints, this will have to be achieved for the continuation of this research project.

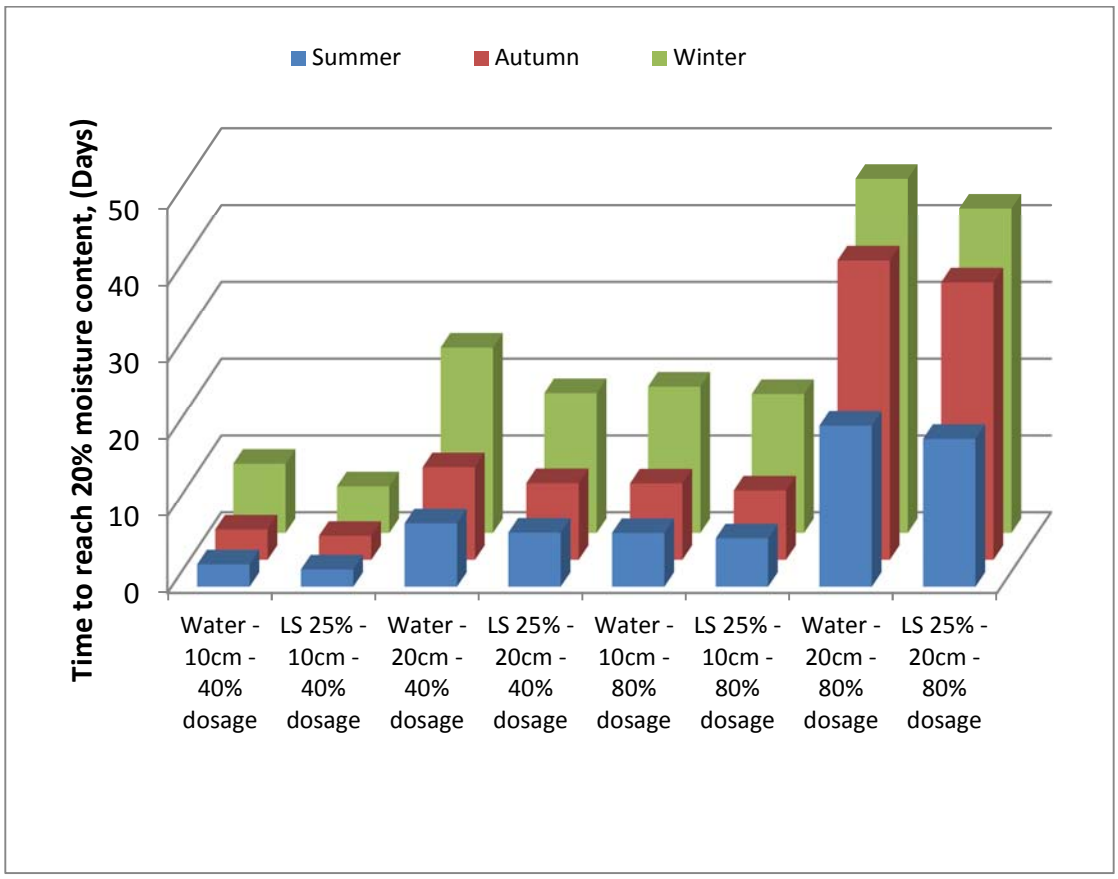


Figure 4.14: Time to reach dry state (20% moisture) for cellulose with varying formulations, sprayed dosage, and insulation thickness.

	Relative decrease in drying times with 25% lignosulfonate additive		
	Summer	Autumn	Winter
10cm - 40% dosage	23%	21%	32%
20cm - 40% dosage	15%	17%	24%
10cm - 80% dosage	10%	9%	5%
20cm - 80% dosage	8%	7%	8%

Table 4.8: Relative decrease in drying times via the contribution of lignosulfonate additives at 28% water concentration.

4.4 FINAL PRODUCT CONSIDERATIONS

4.4.1 Fire resistance measurements

The tested additives used should not interfere with the fire resistance of cellulose insulation provided by the included borate salts and therefore it is necessary to ensure that the fire retardant properties of the material are maintained. There are two types of fire resistance in which cellulose must comply with: smouldering and flame combustion.

4.4.1.1 Smouldering tests

Smouldering is a form of combustion in which no flame is produced it is a slower form of combustion which produces heavy levels of smoke. In order to verify the smouldering resistance, a method defined in the standard ASTM C739 (ASTM, 2006) can be used. The method (see EM21) simply uses a lit cigarette, inserted within the material, to slowly burn the cellulose for two hours, the change in mass due to smouldering combustion is then determined. If this change in mass is higher than 15% the material is considered not smouldering resistant. Sprayed cellulose samples with 25% lignosulfonate and 2% CTAB were tested with regards to smouldering resistance tests and compared to the reference sample sprayed with just water. Both the standard cellulose which has been previously treated with borates and untreated samples were evaluated in order to determine if the additives had any contribution in fire resistance. Results from smouldering tests are shown in Table 4.9. All samples had continuous smouldering combustion during the duration of the tests with no flame created. All samples with borates were under the 15% limit for mass difference in smouldering tests, thus complying with the requirements for insulation materials defined by the standard. None of the samples without borates passed this test, with the additives showing a slightly increased smouldering combustion.

Sample	Mass change (w/w %)
Cellulose reference	5.5%
25% lignosulfonate	6.9%
2% CTAB	5.3%
Cellulose reference (without borates)	60.1%
25% lignosulfonate (without borates)	76.4%
2% CTAB (without borates)	65.1%

Table 4.9: Smouldering combustion test results.

4.4.1.2 Open flame Test

In order to ensure the flame resistance of building materials, a testing method is defined in a European standard EN ISO 11925-2:2010 (ISO, 2002). The test determines the ignitability of a vertically mounted test sample when a small flame is directly applied to its surface. After a period of time, the flame is removed and the specimen is observed to determine whether there is any flame spread to a distance of 150 mm above the point of flame application, within a specified time period (Figure 4.15). The time that the material is exposed to the flame and the observation period is determined according to the material's fire class, ranging from B to E, ranging from difficult to ignite materials to materials which are easily combustible, as defined in 13501-1 (EN, 2010) (see EM22 for more information). In addition, the development of smoke during burning is observed during this period, which is classified in the standard as either s1: no smoke/low smoke production, s2 some smoke production, or s3 high smoke production. Finally, a sample of filter paper is placed under the specimen and, as the specimen is exposed to flame, if the filter paper is ignited by the droplets produced during burning, the material is classified as d0, d1 or d2 according to the presence and persistency of the droplets.

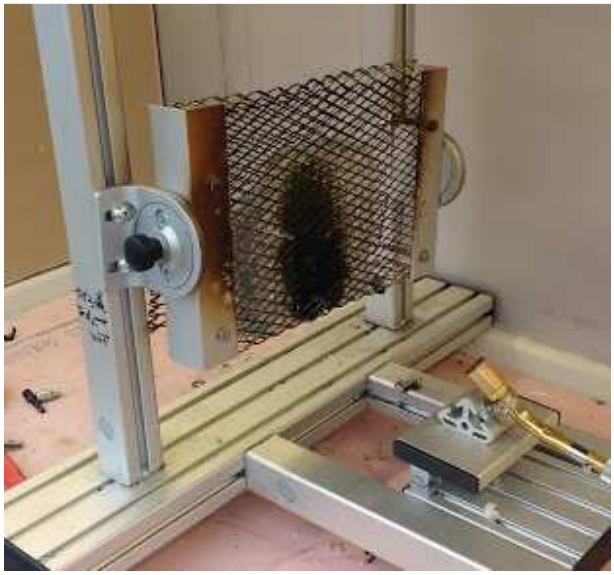


Figure 4.15: Open flame combustion tests setup.

For cellulose insulation the fire resistance classification varies depending on the manufacturer, but many comply with the B-s2-d0 category.

300x90x90 mm^3 sprayed cellulose samples with lignosulfonate and CTAB were studied with regards to flame resistance according to EM22 and compared to reference samples. 3 samples per formulation were tested, with each test measured on a different side. Table 4.10 shows the average measured flame height, the measured change in mass, as well as the observed smoke and droplet behaviour of samples.

Formulation	Flame height, mm	Mass change (w/w %)	smoke level	Ignition of filter paper
Cellulose reference	104 ± 5	7.3% ± 2%	normal	no
25% lignosulfonate	96 ± 6	6.7% ± 1%	normal	no
2% CTAB	110 ± 10	8% ± 2%	normal	no

Table 4.10: Open flame combustion tests.

For samples without borates, flame testing could not be done due to high combustion levels.

All samples complied with the requirement of having a flame height lower than the 150mm threshold specified by the standard. Although variability in samples and the

test method makes it difficult to verify without further results, additives treated with lignosulfonate showed a slightly lower flame height and change in mass than the reference. One cause of this is the higher density of the material, but also the minerals within the lignosulfonate could have a contribution. Smoke was generated from all samples at a similar rate and only a few droplets were produced and the filter paper under the sample did not ignite. From these initial results it's possible to deduce that the fire resistance class of the material is not hindered by the included additives. There were some faults in the testing method however. The sample holding support had a wire mesh to keep it in place, making reading of the flame height difficult and possibly blocking part of the flame. This is exemplified in Figure 4.16.

Using other fire resistance standards to measure such as the single burning item test EN 13823 (EN, 2002), further work on the fire performance needs to be done to fully characterize the material's performance in this regard, with the possibility of included additives being a partial replacement for the borax fire treatment.

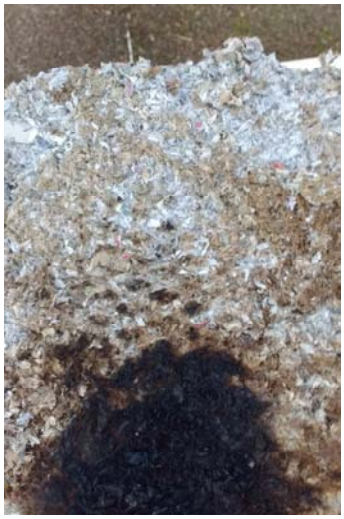


Figure 4.16: Open flame cellulose sample.

4.4.2 Fungal resistance observations

Full fungal resistance tests, like the ones defined in EN 15101 (AFNOR, 2014) are beyond the scope of this work. Chapter 1 has shown how the included borates and other additives already used in cellulose insulation. However, this does not protect the cellulose when it is saturated with water. Some samples with antifungal additives which

were intentionally sprayed at a high dosage of around 250% moisture content showed a yellowish mould on its surface, probably the aspergillus species, after a week of drying (see Figure 4.17).



Figure 4.17: Mould in high moisture cellulose samples.

This should not be an issue if cellulose is properly applied, but can cause problems if there is rain infiltration or pipe leakage in contact with the cellulose. Both lignosulfonate and CTAB addittives have been shown to have some antifungal properties (Madad et al., 2011; Vieira and Carmona-Ribeiro, 2006) As a preliminary test to evaluate the possible influence of the additives on fungal growth, cellulose samples without borates were sprayed at 80% dosage in 50x150x150mm^3 with water, 25% lignosulfonate, 2% CTAB and a high concentration of 10% CTAB. The samples were laid to dry at the standard ambient conditions and then 10g pieces were re wetted at a high moisture content of around 200% moisture, and stored in 95% RH at 25°C using a saturated salt solution, with the intention of creating conditions favourable for mould growth. Figure 4.18 shows untreated, 2% CTAB, 10% CTAB, and 25% Lignosulfonate (LS) samples after 14 days. Discoloration due to fungus can be seen with untreated samples, which is slightly less prevalent with 2% CTAB and 10% CTAB samples show almost no discoloration, resembling the borate treated samples. This could not be determined for lignosulfonate samples due to the colour of the additives.

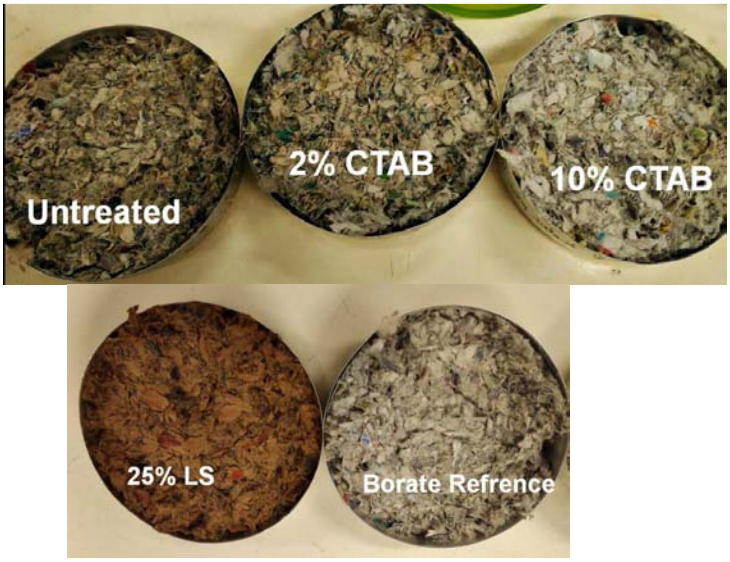


Figure 4.18: Fungal growth discoloration on cellulose samples with different additives.

While the 10% CTAB formulation would not be effective in reducing drying times, they could serve as an alternative to borates as an antifungal additive.

4.4.3 Final product properties and further material studies

Table 4.11 shows the summary of the properties of new formulations with lignosulfonate and CTAB. A “minimal effective dosage” of lignosulfonate at 19% concentration was measured with regards to drying, thermal conductivity and mechanical resistance. If necessary a concentration higher than 25% can be used, although at 40%, heterogeneities in the material can cause issues. In reality, the final effective concentration should be determined via full scale tests and a price evaluation. Despite the fact that the CTAB variant is less effective with regards to drying it could still be used as an additive in the case where maintaining the low thermal conductivity and density of the material is required.

Property	Cellulose reference	19% lignosulfonate	25% lignosulfonate	2% CTAB
Time to reach 20% w/w moisture, (hours) at 60% RH 25°C	51	42	38	45
Density, (kg/m^3)	49.8	54.3	55.6	50.6
Thermal conductivity 10°C, (mW/m.K)	39	40.9	41.1	38.7
Thermal conductivity (25°C mW/m.K)	42.3	44.5	44.8	41.9
Thermal conductivity (40°C mW/m.K)	51	55.2	57.1	52.1
Compressive modulus E, (kPa)	14.1	16.5	16.9	14.3
strain at 5% deformation, (kPa)	0.6	0.9	0.9	0.6
strain at 10% deformation (kPa)	1.3	1.6	1.8	1.2

Table 4.11: Properties summary of cellulose insulation with lignosulfonate and CTAB additives.

From an economic standpoint, the increase in price from the included additive needs to be studied. Considering only material costs, on average around one tonne of cellulose is required to insulate an entire house. If the cellulose is sprayed at the minimal 40%, a 25% lignosulfonate/water ratio translates to 8% lignosulfonate per ton of cellulose so 80 kg per building site. An average price per tonne of cellulose is around 1200€, and for lignosulfonate, prices can range from 300€ to 1000€ depending on the purity, quality and source of the lignosulfonate. Even at the high limit of this range, this only translates to a price increase of 7%. Using the same logic for CTAB, where prices are higher (1000€-10000€ per tonne) this increase is 8.3% due to the low concentration needed (sources: (Alibaba.com, 2016)). The use of these additives also implies an increase in the material used to apply the machine. First a membrane pump needs to be used instead of the standard cellulose spraying pump which connects directly to the water line. A reservoir for storing and mixing the liquid needs to be used. The capacity of the reservoir can range from 100 to 400L depending on the number of times the additive will be mixed with water. For the mixing of the additive, either a portable paint agitator or a concrete mixer (which might be already necessary for the construction site) might be able to be used. The prices on all this extra equipment could vary from 600€ to 2000€ but these are initial investments or could be rented. These increases in cost can be counterbalanced

with the reduction in labour costs and improvement in productivity due to a reduction in drying times for the material.

With regards to production, ideally both the cellulose fibres and the additives should be delivered together to the building site. the manufacture sites of both the provider of cellulose: Univercell (Bordeaux, France), and the producer of lignosulfonate: Tembec (Tartas, France) are within 200 km of each other. Univercell, one of the largest cellulose manufacturers in France, produces 15ktonnes of cellulose insulation, therefore would require 960 tonnes of lignosulfonate/year, assuming complete production would be dedicated to this new formulation. This is only 1.6% of the annual capacity (60 ktonnes/year) of lignosulfonate from Tembec. For the CTAB variation, this becomes more difficult since local providers of the additive are harder to find, with the cheapest providers coming from China. The low concentration required per ton of cellulose can reduce the impact of this issue, since less of the additive is required in the production.

The ecological impact of the final product is also an essential characteristic that needs to be defined. As an initial assessment of the impact of the included additives on the ecological impact of the new insulating material, it's possible to consider the life cycle assessment of cellulose insulation from the work of Zabalza Bribián et al (2011) and integrate the relative impact from the additives with their respective proportions to cellulose insulation. In the case of lignosulfonates, González-García et al. (2011) calculated the environmental impact of this by-product of paper production by considering an economic allocation of a Swedish dissolving pulp mill from a cradle-to-gate perspective. The mass allocation, which can vary according to current market prices of the lignosulfonate by-product, was 1.3% of the ecological impact of paper pulp, another approach could be to quantify using a mass allocation of the final paper pulp produced which was 2.4%. Although no clear data on the life cycle assessment of the CTAB surfactant could be made, data of the ecological impact from the production of an industrial chemical surfactant was taken as an analogue from (Huang, 2008). Table 4.12 shows the approximated ecological impacts, with regards to primary energy demands, and global warming potential, of the novel insulation material formulations.

Material	Primary energy demand (MJ-Eq/kg)	Global Warming Potential (kg CO_2-Eq/kg)
Cellulose insulation	10.4870	1.8310
Cellulose insulation + LS (economic)	10.5159	1.8353
Cellulose insulation + LS (mass)	10.5404	1.8390
Cellulose insulation + CTAB	11.5870	4.8010

Table 4.12: Potential impact of additives on primary energy demand and global warming potential of cellulose insulation.

The influence of lignosulfonate on the ecological impact of cellulose insulation could be considered negligible, thus increasing the interest of the use of the material. Despite the lower concentration of CTAB, its production has a higher impact on both factors. This is primarily due to the high energy requirements for the production of surfactants. In spite of these increases both new formulations still have lower energy demand than traditional insulation materials such as polyurethane foams slabs and mineral wool insulation (Schiavoni et al., 2016). A full life cycle analysis, considering the production distribution and application of the additives should be evaluated in future studies.

There exist other properties of the material that need to be determined. First, as mentioned previously the resistance to mould must be established according to the standard NF EN15101 (AFNOR, 2014). Since the material could be in contact with metal elements the corrosive properties is another factor that is certified for insulation products. The test, also defined in NF EN15101, consists in placing a series of copper and zinc coupons in contact with water saturated cellulose. Other important characteristics to determine for the final product include: Sound absorption determined according to EN ISO 354 and EN ISO 11654, airflow resistivity as defined by EN 29053, a toxicity and dangerous substance report, such as one presented in previous works (Hilado et al., 1979; Morgan, 2006), and odour, which, while not defined in European standards, can be found in ASTM C-1149 (a slight odour for lignosulfonate-treated cellulose was found at high concentrations). A factor not featured in the standards is the fact that the lignosulfonate leaves a residue on the sprayed substrate, leaving small stains on the OSB structure. Since

this part of the building structure is covered and not seen, it should not be an issue, but careful attention should be made to ensure that the stains do not bleed into other parts of the structure.

4.5 CONCLUSIONS

In this final stage of the research project, insulation materials which incorporate recycled cellulose and lignosulfonate and a cationic surfactant were optimized and studied systematically for its subsequent use as a novel building material. In this regard, the concentration of both additives was studied with regards to their influence of drying times and final properties of cellulose insulation. A range of 19%-25% and 2% concentrations for lignosulfonate and CTAB respectively were found to be the most beneficial, although higher concentrations could be used if required, with the disadvantage of having a higher cost, density and thermal conductivity.

In pursuance of the prediction of the drying times of cellulose insulation under real-world dynamic conditions, models of the material's moisture transport were studied. First a simplified empirical model was used in which the moisture content could be predicted using the cellulose insulation's thickness, the sprayed and final moisture contents, as well as the influence of additives. The empirical model provided a strategy in predicting drying times using simple parameters at constant humidity and temperature. Afterwards a complete numerical model was used via the WUFI 5 software so that the combined influence of the lignosulfonate additive with the installed weather, liquid dosage and the thickness of the insulation material could be quantified with regards to the drying time required. The model gave indications on which conditions were the most and least favourable for the use of lignosulfonate formulations. Overall the use of additives would not be recommended when installed at high thickness coupled with high liquid dosage conditions, but the drying times makes these conditions impracticable even without additives. For summer and autumn climates at the minimal spray dosage of 40%, lignosulfonate is recommender at thicknesses higher than 160 mm.

Preliminary studies with regards to the fire performance of the new cellulose insulations were made, which were shown to comply with the required standards. An early study was done for fungal growth, and the included additives could serve as a

partial replacement for the included borax salts. An issue with using the spray additives as either antifungal or fire resistance agents, is that it makes the burden of providing a fire and fungus resistant material more dependent on the cellulose installer and not the manufacturer, increasing the risk of these complications is the material is improperly installed, which might not be suitable for some builders. The final insulation materials with additives show an improvement in drying times and mechanical resistance without greatly affecting its thermal conductivity and density. Lignosulfonate was the most effective in this regard, but CTAB could be used as an alternative when maintaining the material's properties is required. An initial estimation with regards to the relative influence of the additives on the material's price and ecological impact was made, finding them to be negligible in the case of lignosulfonate, with the CTAB variant having a similar price but higher ecological impact in terms of energy demand and global warming potential. Several indications for the characterization of the final product were identified. The corrosion resistance, as well as full fungal resistance tests are necessary properties that need to be defined for the validation of the material. Other properties include: sound absorption, airflow resistance, toxicity, and odour testing, amongst others. Finally, full scale onsite tests need to be done at a pilot construction site to verify the effectiveness of the additives under full scale conditions and in comparison to the numerical models.

GENERAL CONCLUSIONS AND PERSPECTIVES

The challenge of sustainability in the building sector needs comprehensive examination. Building projects require innovative materials that not only improve its properties with regards to energy consumption, but also reduce the ecological impact of the building using building components with natural and/or recycled raw materials, all while taking into account practical factors such as price, availability, and impact on health. Despite its performance in improving thermal comfort and its eco-friendly nature, cellulose is not a prevalent building material when compared to traditional materials such as glass wool. The main issue with the material resides from its installation in building envelope enclosures. The dry blown method is prone to settling which causes voids that create thermal bridges, while the wet process can require a long time to dry, depending on the conditions in which it is applied, which slows down construction schedules. As a result of the work of this thesis, a better understanding on the performance and properties of cellulose insulation applied was achieved, and a new formulation of the material was found in which the drying and mechanical performance of the material was improved. This contributes to the attractiveness of the material and technique for its use in future building projects, thus having a positive influence on the improvement of sustainability in the building sector.

An analysis of the state of the art on cellulose insulation has shown its beneficial properties including its thermal conductivity, low density, moisture buffering potential and airtightness. There is, however, a void in the literature with regards to the wet spray technique. While the problem with the material's drying has been established, no information was available on how sprayed moisture affects its properties. The results of this analysis helped shape the strategy for the study on the properties of cellulose insulation installed via the wet spray process, and the means of which the drying problem could be solved via the use of additives. The potential additives which could improve the

properties of wet spray cellulose were categorized, with a focus on biobased binders and surfactants, with the purpose of maintaining the eco-friendly properties of the insulation material. Despite the several examples of the use of biobased binders with natural fibre composites, the specific conditions for the wet spray process made it difficult to determine the specific feasibility of the use of additives with cellulose insulation, requiring further investigation.

Characterization of wet spray cellulose insulation required an initial study on the properties of fibres, and their physical and chemical properties. Differences between the composition, granulometry, degree of refining, and water retention between two types of cellulose insulation fibres translated directly into differences between drying times of the sprayed insulation, although direct quantification of the degree of influence for each factor could not be defined. Isothermal dynamic absorption testing and the quantification of different type of water within the fibres via differential scanning calorimetry proved to be useful tools in studying the drying rate of sprayed cellulose, where the proportions of bound and unbound water had an influence on the rate of evaporation of water within the fibres.

During the wet spraying of cellulose insulation, the initial moisture dosage can vary depending on the applier. It was therefore necessary to determine how this dosage can affect the final properties of the material. An increase in the density, compression resistance and thermal conductivity was found for increasing installed moisture for cellulose insulation. This increase could be partially explained by the compaction of the material from the increase in water pressure, but when comparing with dry compacted samples, it was evident that other factors such as the pore closure and the hardening of fibres during drying also played a role. The properties of cellulose installed with the minimal 40% moisture content provided a comparative reference for the formulations of cellulose with included additives.

The types of additives considered for the use with cellulose insulation were biobased binders and surfactants. For binders, one of the limitations for their use with the wet spray method was viscosity. The pump and spray mechanisms would only spray liquids of around 40 mPa.s, thus limiting the concentration of additives to be used. A compromise had to be made in where the concentration ranges for the biobased binder

candidates were varied in order to have an adhesive with maximal adhesive strength, but was still under the viscosity threshold. Some of the screened binders, when diluted to a concentration which was sprayable, presented low adhesive strength using small scale tensile strength tests, severely limiting number of possible biobased binders which could be incorporated with cellulose insulation. Compression tests on sprayed cellulose insulation with biobased binders were made. Some of the sprayable binders did have a positive contribution on mechanical resistance of cellulose insulation, while others showed the same as with cellulose sprayed with just water. The additives also had an impact on density and thermal conductivity similar to the contribution of increased water dosage. The addition of biobased binders could not have a direct contribution in the reduction of drying times for cellulose insulation, mainly due to the reduced flow from the viscous liquid and the formation of a crust which impeded drying. The strategy in which cellulose was sprayed with less than the minimal liquid dosage, compensated by increase in mechanical strength from binders was not fruitful either. The only biobased binder which showed a significant improvement in drying was lignosulfonate, due to the decrease in initial moisture content from high concentrations of the additive.

A range of surfactants were also tested, with varying charge and hydrophilic properties. It was initially thought that the reduction in surface tension of the sprayed liquid would improve the drying of the water, within the fibres, but most showed little to no change in drying times. The only effective surfactant was CTAB, mainly due to its cationic nature. The interactions between the positively charged CTAB surfactant and the negatively charged cellulose fibres induced a slight reduction in drying times. Both lignosulfonate and CTAB were considered suitable additives due to their contribution to both mechanical resistance and drying times, with relatively low impact on density and thermal conductivity.

The study on the effect of CTAB and lignosulfonate concentrations on the properties of cellulose insulation yielded the optimal additive/water ratios of 19-25% for lignosulfonate, and 2% for CTAB. In order to account for different conditions in which wet sprayed insulation could be applied, such as installed moisture, insulation thickness, and the use of additives. An empirical model was applied for cellulose drying at constant humidity and temperature conditions. The model gave a good indication of the impact of

each individual parameter on drying times. A more complete simulation, taking into account the properties of the material was done using numerical modelling software. By considering real world climate and material conditions, it was possible to determine at which conditions the lignosulfonate additive was the most and least effective. It was found that the decrease in drying times from the additive would be negligible for low thicknesses, low sprayed dosage, in summer conditions, and for high thicknesses, high sprayed dosage during winter and autumn conditions.

Preliminary studies on the fire resistance of the new formulations were made. They were found to be compliant with the requirements for both smouldering and flame resistance, but only with the included borate salts already used. CTAB surfactants had a contribution on the fungal resistance of the material but only at high concentrations. Partial replacement of the included borate salts with the tested additives for both fungal and fire resistance are a possible added benefit that needs to be studied further.

For the continuation of this work, a full scale drying test, directly in a construction site is envisioned. Moisture meters within the material after it is sprayed, coupled with ambient humidity and temperature sensors will allow the continuous monitoring of the drying of the new insulation material and a confirmation of the numerical model. Further work also needs to be done on the compliance of the material with national and international standards for insulation materials.

Beyond the industrial objective of the development of the innovative insulating material, further research on cellulose insulation, encompassing different scientific areas, could be envisioned. While in an industrial scale the optimization of the physical chemical properties of the fibres is not feasible, an in-depth study on the optimization of the chemical composition and physical properties of the fibres could yield results that are applicable in the production of paper or materials incorporating lignocellulosic fibres.

From a fluid mechanics standpoint, the spreading behaviour of the sprayed liquid, the formation of droplets and their spread on the cellulose fibres as a function of viscosity is a possible topic of further study. Moisture flow through porous media is a subject in which the theory and experimentation is continuously evolving, which could be applied to the study of drying of cellulose. The fibrous network in which moisture flows from the cellulose interior to its surface could be studied via tomography, for example.

The mechanical behaviour of sprayed cellulose insulation is another remarkable problem to examine. Since the material adheres to all the borders of the wall cavity, it is subject to compressive, tensile and shearing forces from its own weight. These forces are dynamic in nature since as the material dries, the applied weight decreases but the material's resistance strengthens. Also, once the material is dry the changes in humidity moisture absorption and desorption throughout the makes the material's mechanical performance cyclic, requiring creep strength testing. This problem is complicated further by the fact that cellulose insulation could be sprayed in different configurations, not only the different dimensions of the wall cavities which directly affect the weight of the material, but also the different wall materials (OSB, fibreboard, gypsum board, etc.) affect its adherence.

Materials such as cellulose insulation are essential components for the reduction of the environmental impact of the building sector, yet their integration in building projects is still less favourable than traditional materials. It is only through iteration, innovation, and research that building materials that incorporate natural and recycled fibres will become more prevalent, the norm and not the exception.

EXPERIMENTAL METHODS

EM01 DETERMINATION OF MOISTURE CONTENT

For determination of moisture content of cellulose samples, the gravimetric method was used. 1-3g of the sample was weighed in its humid state, and then dried in an oven at 103°C and weighed again the moisture content is then:

$$MC(\%) = \frac{m_{wet} - m_{dry}}{m_{dry}}$$

To measure the drying of CFI, the mechanical testing samples were weighed daily in order to measure the evaporated water until equilibrium moisture conditions at 60% RH (a mass variation of less than 1% in 24 hours) were reached (m_{eq}). The equilibrium moisture content was then determined by taking 3g of fibres from the ambient dried sample and drying them at 100°C for two hours. Using the mass of the samples and the equilibrium moisture content, the initial moisture content as well as the drying of the material at 24 hour intervals was determined.

The total moisture content is then

$$MC(\%) = MC_{sprayed}(\%) + MC_{eq}(\%)$$

With

$$MC_{sprayed}(\%) = \frac{m_{wet} - m_{eq}}{m_{dry}}$$

m_{dry} and $MC_{eq}(\%)$ were determined via the oven dried method.

EM02 MACROSCOPIC OBSERVATION OF CELLULOSE FIBRES

The cellulose fibres were observed using a Nikon SMZ 1500 Multizoom microscope. Fibres were placed between transparent plates under the microscope and the lighting, focus and zoom of the microscope was adjusted until the fibre surface could be observed. Images were taken at 10x and 20x zoom.

EM03 SCHOPPER REIGLER FREENESS TESTING

For Schopper Reigler measurements, a standard tester was used. The procedure, adapted from ISO 5267-1:1999, is as follows. Cellulose fibres were mixed with water to form a suspension of 0.2% consistency. The suspension was then subjected to disintegration via a disintegrator at 6000 revolutions. The temperature of the pulp was adjusted to be around 20°C. 1L of the pulp suspension was then slowly poured into the drainage chamber of the Schopper Reigler apparatus. Once 5s had passed, the sealing cone was raised and the pulp suspension was allowed to flow through the apparatus. When all of the liquid had stopped flowing through the apparatus, the volume of liquid in the recipient cylinder was measured. The SR value is then:

$$SR° = \frac{1000 - Vcylinder(ml)}{10}$$

Tests were repeated three times with different pulp suspensions for each sample.



Figure E1: Schopper Reigler freeness tester

EM04 TAPPED DENSITY

Tapped density was done through a Granuloshop Densitap ETD-20 (France) measurement apparatus. 3-5g of loose cellulose insulation fibres were first weighed (m_0) and then inserted into the apparatus. Careful attention was made to ensure that the fibres did not stick to the measurement cylinder walls. The bulk height e_0 was then determined.

The bulk density ρ_0 was determined as the ratio of m_0/e_0 in g/L. the material was then subjected to a series of mechanical taps. A sequence of 100, 300, 500, and 1250 taps were used until the material's height didn't vary anymore. The height e_{tapped} was measured and tapped density ρ_{tapped} was determined as m_0/e_{tapped} . The compressibility index CI was then:

$$CI(\%) = \frac{e_0 - e_{tapped}}{e_0}$$



Figure E2: Tapped density apparatus.

EM05 PARTICLE SIZE DISTRIBUTION

The particle size distribution of the material was determined by first separating around 10g of fibres using compressed air. The fibres were then weighed and put through a series of sieves with decreasing whole diameter. The sieve was covered and compressed air was blown to make the fibres separate and pass through the sieve. The fibres that did not pass through the sieve were weighed and its proportion to the total mass of the sample was determined. $m_{sieve}/m_{total.}$ This value was added cumulously for each successive sieve until all the material was measured, adding to 100%. The material that passed through the sieve passed to a smaller sieve and the process was reported. The sieve diameter sizes used were 4mm, 2mm, 1mm, 0.5mm, and 0.25mm.

EM06 ADF/NDF ANALYSIS

This method allows determining the proportions of lignin, cellulose and hemicelluloses of fibres. The following reagents were used, according to the method described by Van Soest and Wine (1967).

Neutral detergent solution:

Sodium borate decahydrate (Na2B4O7 - 10 H2O) 6.81 g

Disodium ethylenediaminetetraacetate (EDTA, C10H14N2Na2O8) 18.61 g

Sodium lauryl sulfate neutral (C12H25NaO4S) 30 g

2-ethoxyethanol (Ethylene glycol monoethyl ether, Cellosolve, C4H10O2) 4.56 g

10 ml Disodium phosphate anhydrous (Na2HPO4)

Distilled water 1000 ml.

Sodium borate and disodium EDTA

n-octanol (C8H18O) octilic alcohol

Sodium sulfite anhydrous (Na_2SO_3) 4.

Acetone

Acid detergent solution:

Cetyltrimethylammonium bromide technical grade (C19H42BrN) 20 g

Sulfuric acid 1 N (H2SO4, 49.04 g/l) 1 l

Dissolve tensioactive into acid while stirring to promote dissolution.

n-octanol (C8H18O) octilic alcohol.

Acetone.

The procedures for both methods are as follows:

Procedure for NDF determination (Neutral detergent fiber)

1. Grind the air dried sample to pass 1 mm screen.
2. Weigh in a crucible 1 g of grinded sample with 1 mg approximation.
3. Add 100 ml of neutral detergent solution at room temperature into crucible with 0.5 g of sodium sulfite and some drops of n-octanol.
4. Heat to boiling and reflux 60 minutes from onset of boiling.
5. Filter and wash 3 times with boiling water, then twice with cold acetone.
6. Dry 8 hours at 105 °C and let cool in a desiccator.
7. Weigh.
8. Calculate neutral detergent fiber: NDF % = (weight of crucible + weight of residue) - weight of crucible / weight of sample x 100. Neutral detergent solubles: NDS % = 100 - NDF %.
9. Ash in a muffle at 550 °C 2 hours and let cool in a desiccator.
10. Weigh.

11. Calculate ash insoluble in neutral detergent: loss on ashing / weight of sample x 100.

    Procedure for ADF determination (Acid detergent fiber)

1. Grind the air dried sample to pass 1 mm screen.
2. Weigh in a crucible 1 g of grinded sample with 1 mg approximation.
3. Add 100 ml of acid detergent solution at room temperature and some drops of n-octanol.
4. Heat to boiling and reflux 60 minutes from onset of boiling.
5. Filter and wash 3 times with boiling water, then twice with cold acetone.
6. Dry 8 hours at 105 °C and let cool in a desiccator.
7. Weigh.
8. Calculate acid detergent fiber: ADF % = (weight of crucible + weight of residue) - weight of crucible / weight of sample x 100.
9. Ash in a muffle at 550 °C 2 hours and let cool in a desiccator.
10. Weigh.
11. Calculate ash insoluble in acid detergent: loss on ashing / weight of sample x 100.

EM07 DYNAMIC VAPOUR SORPTION (DVS)

Sorption isotherms were made by dynamic vapour sorption apparatus DVS Advantage from Surface Measurement Systems Ltd (London, United Kingdom). Loose fibre samples were placed into an aluminium sample holder connected to a microbalance. The samples were then subjected to a series of relative humidity variations from 5% to 95%, with 10% intervals, at a constant temperature of 25°C. The variations in mass due to moisture adsorption and subsequent desorption were then plotted against relative humidity.



Figure E3: DVS measurement apparatus.

EM08 WATER RETENTION VALUE

The water retention value (WRV) of paper was determined through a method inspired by ISO 23714. Around 5g of fibres were suspended in water for 24 hours. The sample was removed from the water and were let air dry in a wire mesh to remove water present in the fibre surface. The fibres were then placed in a centrifuge tube with a cloth mesh. The samples were then centrifuged at a speed of 3000g for 5 minutes. The fibres were then and weighed ,$m_{centrifuge}$. Then the fibres were then dried at 100°C for 24 hours and re weighed m_{dry}. The WRV is then:

$$WRV = \frac{m_{centrifuge} - m_{dry}}{m_{dry}}$$

EM09 BOUND WATER DETERMINATION.

The bound and unbound water contents were determined according to the method described in Nakamura et al. (1981). The method uses differential scanning calorimetry (DSC) to freeze the wetted fibres and measure the melting enthalpy as the frozen fibres reach 0°C. In these melting curves two peaks can be observed: one corresponds to the frozen bound water and the other to free water. The remaining water is considered to be the non-freezing bound water. 50g of untreated Univercell loose fibre samples were

moistened at around 100% moisture content then left to dry at 60% RH and 25°C. Around 5mg of fibres were then placed in an aluminium DSC crucible and sealed. DSC measurements were made from -30°C to 15°C using a DSC 3 apparatus from Mettler-Toledo (Columbus, USA). An example melting curve is shown in Figure E4, with the integral of both peaks highlighted. As the fibres dried, samples were placed in the DSC apparatus and the melting enthalpies were measured from decreasing moisture content. The melting peaks were then integrated to determine bound and free water content as a function of total water. The melting enthalpy for the type of water was divided by the known melting enthalpy for water, 334 J/g, to give proportions of each type, with the remaining water attributed to nonfrozen bound water. The amount of water in a melting peak was calculated according to: (Weise et al., 1996)

$$X_{water}\ (\%) = \frac{\Delta H_{peak}}{H_f \cdot m_{dry}}$$

Where: ΔH_{peak} is the energy transferred according to the peak melting peak, $H_f$the specific heat of fusion of water (334 J/g) and m_{dry} is the mass of solids (g).

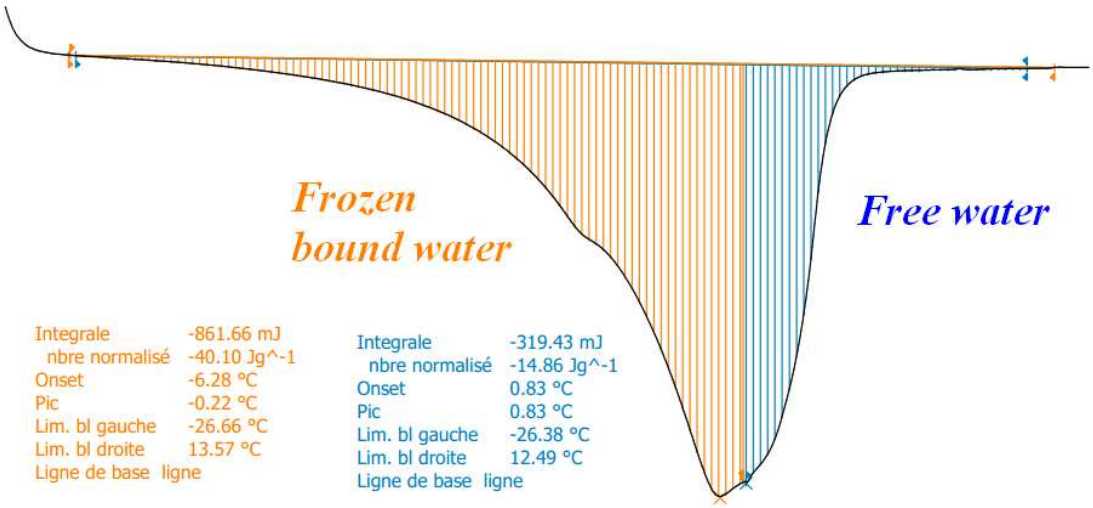
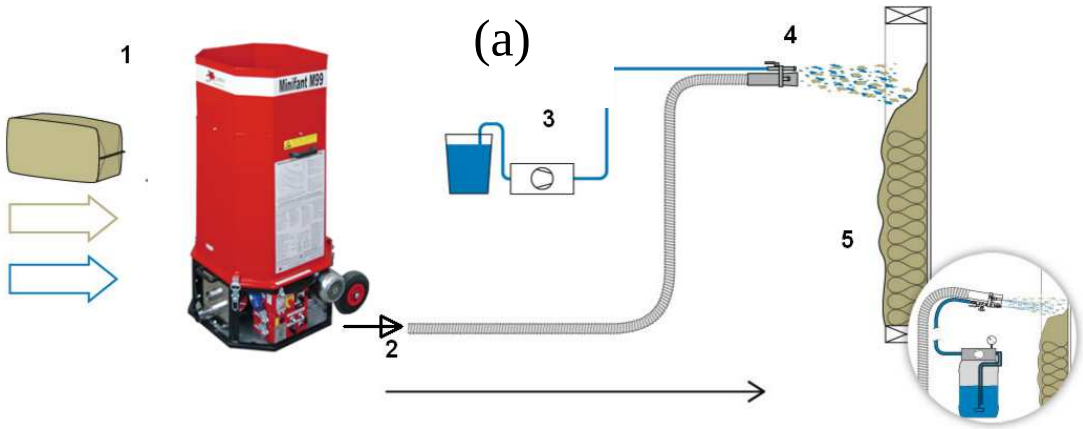


Figure E4: DSC melting curves.

EM10 SPRAYING OF WET SPRAY CELLULOSE SAMPLES

Cellulose fibre insulation was sprayed with a Minifant M99 blowing machine (Figure E5a) from X-Floc (Renningen, Germany). The blower was coupled with a COSPET membrane pump (Poviglio, Italy) (Figure E5b) to spray the water. The pump was connected via a hose to two nozzles on both sides of the main blowing hose (Figure E5c). The spraying procedure as shown in Figure E4a is as follows: (1) the compacted fibres are fed to the blowing machine, which first separates them so they can be blown (2). For the wet spray method, a separate pump, connected to the blowing hose, pumps water from a 50L bucket (3) which is then atomized through nozzles at the same time the fibres are blown into the cavity (4). The excess material is then removed via an electric wall scrubber (5), (Figures E5d and E7). The blowing machine allows the control of material flow via two methods: the opening of the material feed gate and the air pressure ranging from values of 1 to 9, 9 being the feed gate completely open, and air pressure ranging from 0 to 100% . Too little air pressure can cause excess water to be applied, and too much air pressure creates dust and can create a force that makes the material detach from its cavity. Unless noted otherwise, most samples were sprayed at a configuration of a feed gate of 4 and an air pressure of 40%, which provided a sufficient material flow rate

to ensure proper applying of the material. This corresponds to an air flow rate of around 140 m^3/h and a material flow rate of 220 kg/h. In order to vary the initial moisture content of the samples, the pumped spray water pressure varied from 6 to 20 bars. For cases where an exaggerated water dosage was applied, the air pressure/ feed gate configuration was 20/8 with a water pressure ranging from 10 to 20 bars.



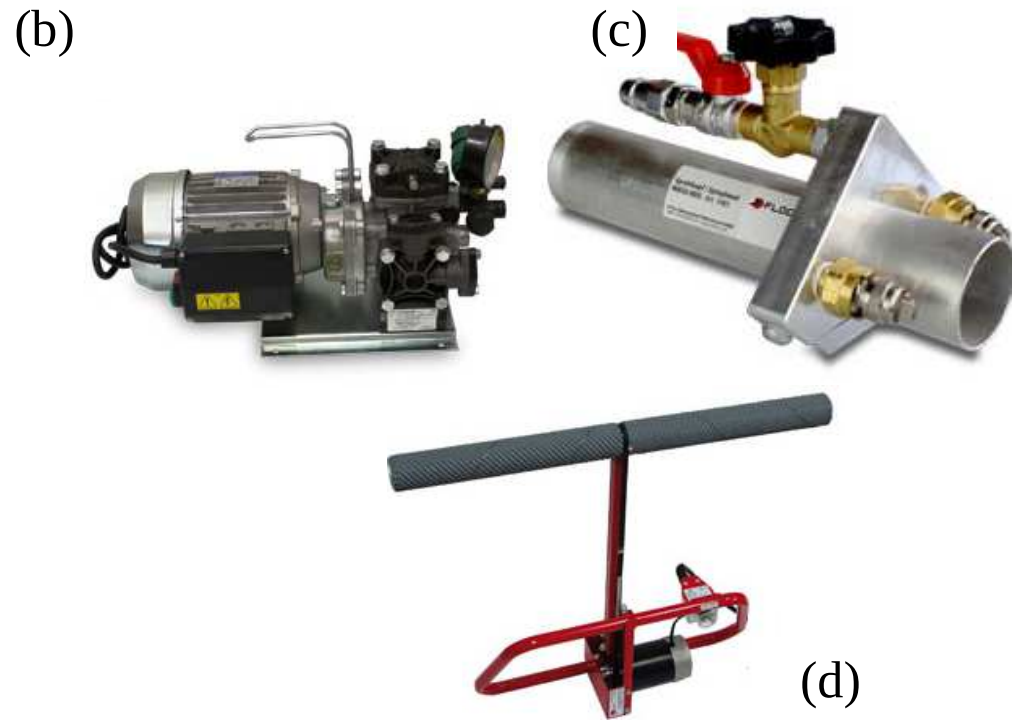


Figure E5: (a) Cellulose blower and wet spraying procedure (b) Wet spraying water pump (c) blowing hose with water nozzles (d).



Figure E6: Cellulose wet spraying.



Figure E7: Excess material removal.

The fibres were blown into wooden moulds of dimensions: 300x300x90mm^3 for drying and mechanical tests. The moulds were fitted with a removable 2.5mm rigid cardboard base covered with a coat of vinylic glue. Once the insulation sample was dry, it was carefully unmoulded and another 2.5mm cardboard was glued to the other side of the sample. Once the glue dried, the sample was then cut into nine 100x100x90 mm^3 pieces via a circular saw. The use of cardboard served two purposes: to ensure that the

100x100mm² samples have good contact with the compression plates and that they were cut evenly without loss of the material. Thermal conductivity samples were projected on 150x150x50mm^3 moulds with a removable top for unmoulding. Once dry, samples were carefully separated from the moulds using a trowel. All samples were stored in a climactic chamber at 25°C and 60% relative humidity (RH).

In order to have samples with variable density but similar sprayed moisture contents, the 90mm thick compression testing moulds were adapted to have 120 140 and 160 mm thickness using woodboard. The thicker samples were sprayed at the minimal 40% moisture and dried at similar conditions of the other samples. Once dry a metal plaque was placed on top of the sample, with iron weights ranging from 0.5 kg to 4 kg were placed upon the plate to compact the material. The material was compacter for 24 hours to ensure it would not regain its density once the plate was removed. This would ensure that the samples would have higher density, but similar thickness (90mm) and initial applied density. The same was done with thermal conductivity samples whose moulds were adapted from 50mm to 70mm and 90mm.

EM11 COMPRESSION TESTING

Compression tests, adapted from the standard NF EN 826, on the cut 100mm x 100mm x 90mm samples were made by means of an H5KT universal testing machine from Tinius Olsen (Surrey, England) equipped with either 100N or 500N sensors, depending on the material's resistance. Steel 100 x 100 mm² compression plates ensured direct contact with the samples. Compression was done at a speed of 10 mm/min. A pre-charge of 1 N was applied to ensure direct contact with the sample. Due to the fact that the material is highly compressible, no fracture or rupture was detected so instead the measured stress at 5% and 10% strain was logged for each sample. Compression tests stopped once 20% strain was reached. Since each sample had slight variations in dimensions of the order of ± 2mm, they were measured in order to accurately calculate their respective stress and strain.

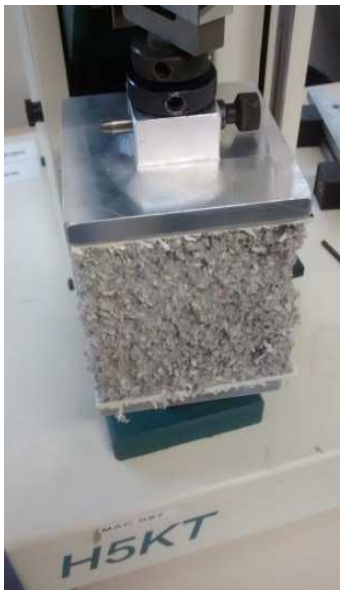


Figure E8: Compression tests.

EM12 THERMAL CONDUCTIVITY MEASUREMENTS

Thermal conductivity measurements were done according to the standard NF EN 12667. Samples of 150x150x50mm^3 were placed in a λ-Meter EP500e guarded hot plate apparatus from Lambda-Messtechnik (Dresden, Germany). Foam insulation was used around the samples to ensure the contact plates measured only the 150 x 150 mm² surface of samples. The contact plates applied a pressure of 50 Pa to ensure direct contact with the samples without compacting them. Measurements were made at 10 and 25° C average temperature with a temperature difference of 10°C between plates. Steady state was assumed to be reached when the value of thermal conductivity varied less than 1% in 60 minutes.



Figure E9: λ-Meter EP500e apparatus.

EM13 BIOBASED BINDER PREPARATION

A wide range of polysaccharide binders and gums were tested. Citrus pectin powder, guar gum, xanthan gum, pregelatinized corn starch, and sodium alginate, were obtained from Kalys (Bernin, France)., produced from corn starch of the brand GLUCIDEX® IT 19 were obtained from Roquette (Lestrem, France). In order to have a comparison pont with synthetic binders, a commercial of the brand Unibond 1C was obtained from Unibond (Cheshire, UK), its solids content is 39%.

Phenolic biobased adhesives were among the addittives tested. Low sulfonate lignin was obtained from Sigma Aldrich (Brussels, Belgium), lignosulphonates were obtained from Tembec (Tartas, France).

Soy protein isolate was obtained from Activ'Inside (Libourne, France) it was tested in neutral water and in 10^{-4}M/L Sodium hydroxide (NaOH) buffer with a pH of 10.

Cationic mimosa tannin of the brand Tanfloc was obtained from Tannac (Hamburg, Germany). Cationic corn starch of the brand LAB 4202 was obtained from Roquette. This binder was in liquid form, with 42% solids content, which was taken into account for successive dilutions. Chitosan powder, also cationic, was obtained from Kalys, and mixed with 1% acetic acid ($C_2H_4O_2$) solution (pH=5).

Powdered binders were diluted at variable concentration with around 50g of distilled water and mixed with a magnetic stirrer for one minute and let to settle for ten minutes to avoid thixotropic loss of viscosity. Since some binders are more viscous than others, the range of concentrations tested varied for each binder in order to attain the proper pumpable viscosity for some binders. In order to determine the maximum allowable viscosity, a preliminary test with the spraying equipment and two biobased binders: pregelatinized wheat starch and apple pectin. Binders were mixed with water, staring at 10% concentration, and gradually decreasing until the liquid was pumpable. It was found that for 1.5% pectin and 1% starch concentrations, binders were sprayable at a rate similar to that of pure water. This will further be optimized for other binders in section 3.5.3

EM14 VISCOSITY MEASUREMENTS

Rheometric analysis on biobased binder solutions was carried out with an Anton-Paar (Graz, Austria) MCR302 rheometer with a cone-plane geometry (D=25mm) at 25°C. 2 ml of each of the prepared binder solutions was placed under the plate using a micropipette. The cone was then lowered, with the excess liquid removed. Measurements were setup at a linear incremental step shear rate from 0.1 to 20 s^{-1}, with 20 points of measurement. Afterwards, if a shear thinning behaviour was observed, the same binder solution was tested using a power step rate with 20 points from 0.1 to 3000 s^{-1}.



Figure E10: viscosity measurement apparatus.

EM15 TENSILE STRENGTH TESTS

Used newspapers recovered from the cellulose insulation production line from SOPREMA. The newspaper was cut into 20mm x 50mm strips. The single strips were weighed then dipped into the binder solutions (the same used for viscosity measurements). The covered strips were weighed and excess liquid was removed using tissue paper in order to have around 200mg of liquid on all paper samples. Samples were then laid to dry at 60% RH and 25°C for 24 hours. The dry paper strips were then placed on the same H5KT universal testing machine from Tinius Olsen (Surrey, England) equipped with a 100N sensor and special tensile strength clamps. The paper was fitted between the clamps 30mm apart. The paper strips were then subjected to tensile tests at 10mm/minute and the maximum tensile force at fracture; F_{max} was measured for each sample.

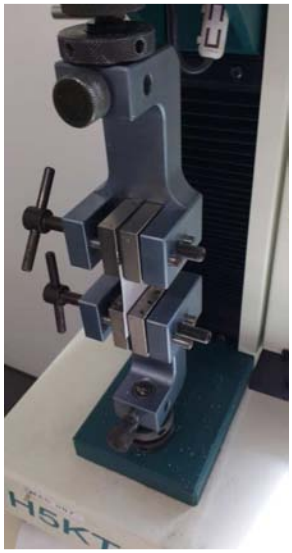


Figure E11: Tensile strength tests.

EM16 SURFACE TENSION MEASUREMENTS

Surface tension measurements for surfactant solutions were made using a 3S tensiometer from GBX (Dublin, Ireland) via the Wilhelmy plate method. Measurements were performed at room temperatures. All glassware and the platinum plate were rinsed with ethanol prior to each measurement, the platinum plate was burned using a lighter to remove traces of any residual material. The solution is placed on the glass recipient. The Wilhelm plate is subsequently lowered slowly until it is under the surface of the liquid. It is then raised until the plate is slightly over the surface. The measured force F is then divided by the length of the plate: 40mm, to give the surface tension in mN/m.

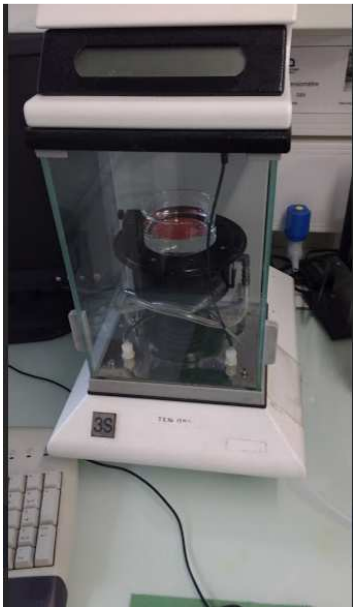


Figure E12: Tensiometer.

EM17 EMPIRICAL DRYING MODEL OF CELLULOSE INSULATION

Using equation 4.2, the model was fitted with experimental results by optimizing the time constant t_c. This was done by calculating the root mean squared error between the predicted values and the experimental measured drying curves using the equation:

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(\widehat{w_i} - w_i)^2}{n}}$$

With $\widehat{w_i}$ and w_i the experimental and calculated values for the drying moisture contents after each time step, and n the number of measurements.

The drying constants were then optimized by reducing the root means square for the drying times using the solver function in Excel, which uses the generalized reduced gradient algorithm, to find the value of t_c which minimises the RMSE for each case.

EM18 SPECIFIC HEAT CAPACITY WITH DIFFERENTIAL SCANNING CALORIMETRY

Fibre specimens ranging from 10-15 mg ware taken from sprayed cellulose samples after drying, to equilibrium moisture content in a climatic chamber. Samples were placed in 100µl crucibles and inserted into a DSC 3 apparatus from Mettler-Toledo (Columbus, USA). To measure specific heat capacity, the temperature modulated DSC

TOPEM technique was used. The method and its theory is detailed in Fraga et al. (2007). Using a time series of stochastic (random) temperature pulses of different durations, quasi-static specific heat capacity can be measured by integrating the pulse response curve. Specific heat capacity of samples was measured using a temperature range of 15 to 35 °C.

EM19 MOISTURE DIFFUSIVITY COEFFICIENT VIA CAPILLARY SORPTION

In order to determine the diffusion coefficients using equation 4.7, as defined by (Künzel, 1995) the short term water absorption coefficient is calculated using a method based on the standard BS EN ISO 15148:2002 (British Standards Institute, 2002). In these tests, the loose cellulose is placed in a 150mm diameter cylindrical recipient at a height of 200 mm with a wire mesh covered with a polyester net at the bottom. The bottom part of the material is in contact with a water recipient (See Figure E13).

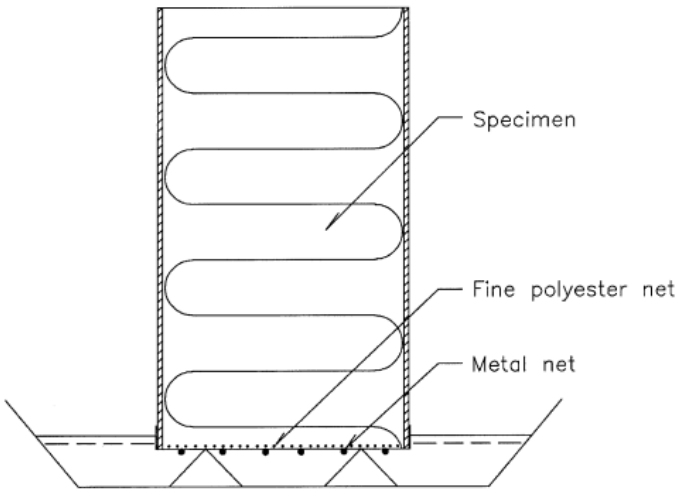


Figure E13: Capillary sorption measurement setup (Hansen et al., 2001)

Then the capillary water flow is measured as kg/m² of absorbing surface. The mass change is weighed periodically and plotted against the square root of the weighing times. From the slope of the initial moisture sorption, the water absorption coefficient is determined (see Figure E14).

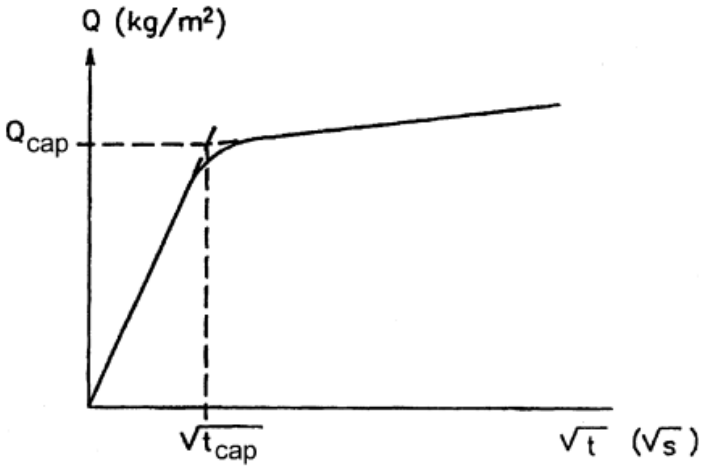


Figure E14: Water absorption coefficient determination (Hansen et al., 2001)

EM20 NUMERICAL DRYING MODEL USING WUFI SOFTWARE

In order to define the drying model, the WUFI software requires the definition of the following category of parameters:

Component parameters: the wall section and layers are defined, with the wall assembly layers and materials, orientation, inclination of the wall, the surface transfer coefficient, and the initial conditions,. It is this section where the initial moisture dosage (40 or 80%), and the insulation thickness (10 or 20cm) is defined.

Control parameters: the calculation period is defined, which in this study was either the month of January, October or August or the time step of 1 hour was chosen. Both simultaneous moisture and heat simulations were applied.

Climate parameters: here a database of climate conditions can be chosen, In this case the climate file from Bordeaux 2008, provided from Météo France, was used. For fitting of diffusivity curves the conditions of the climactic chamber (25°C 60% RH) was used.

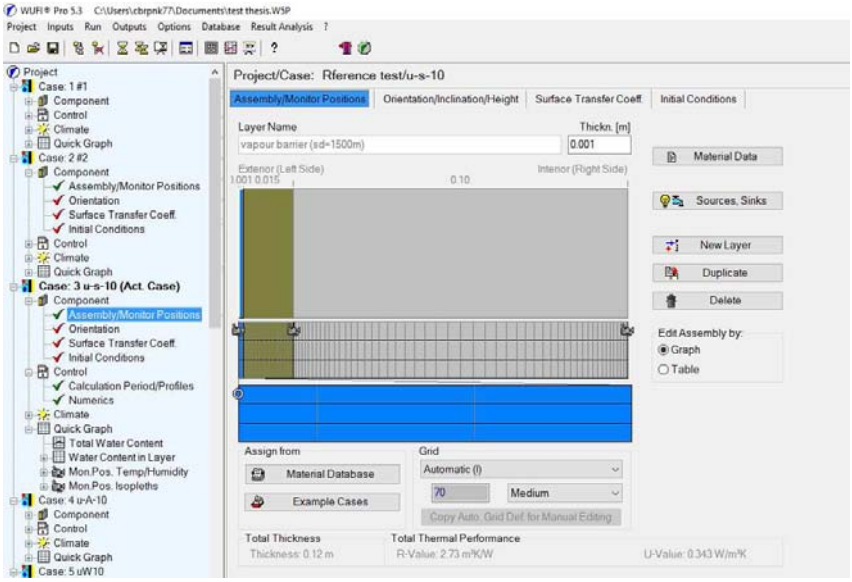


Figure E15: WUFI 5 software UI.

Simulation run	Material type	Sprayed dosage	Thickness	Weather profile
1	Reference water	40%	10 cm	Summer
2	Reference water	40%	10 cm	Autumn
3	Reference water	40%	10 cm	Winter
4	Reference water	40%	20 cm	Summer
5	Reference water	40%	20 cm	Autumn
6	Reference water	40%	20 cm	Winter
7	25% lignosulfonate	40%	10 cm	Summer
8	25% lignosulfonate	40%	10 cm	Autumn
9	25% lignosulfonate	40%	10 cm	Winter
10	25% lignosulfonate	40%	20 cm	Summer
11	25% lignosulfonate	40%	20 cm	Autumn
12	25% lignosulfonate	40%	20 cm	Winter
13	Reference water	80%	10 cm	Summer
14	Reference water	80%	10 cm	Autumn
15	Reference water	80%	10 cm	Winter
16	Reference water	80%	20 cm	Summer
17	Reference water	80%	20 cm	Autumn
18	Reference water	80%	20 cm	Winter
19	25% lignosulfonate	80%	10 cm	Summer
20	25% lignosulfonate	80%	10 cm	Autumn
21	25% lignosulfonate	80%	10 cm	Winter
22	25% lignosulfonate	80%	20 cm	Summer
23	25% lignosulfonate	80%	20 cm	Autumn
24	25% lignosulfonate	80%	20 cm	Winter

Table E01: Experimental run design parameters.

EM21 SMOULDERING TESTS

Smouldering combustion tests for cellulose insulation were adapted from the standard ASTM C739 (ASTM, 2006). Cellulose samples of 200x200x60 mm^3 are sprayed and left to dry in a climactic chamber. Once dry they are weighed and placed in a metallic recipient. In the test a lit cigarette is inserted, lit side up within the material. The cigarette is left to burn for two hours or until smouldering has ended. After the test period, the remaining material and any residue are re weighed. If the weight loss does not exceed 15%, the material is considered resistant to smouldering combustion.

EM22 FLAME IGNITABILITY TESTS

The method for this test was adapted from the standard EN ISO 11925-2.The test determines the ignitability of a vertically oriented test specimen when exposed to a small flame, at the edge of the specimen. The burning behaviour of the specimen is observed for flame spread, and the occurrence of burning particles and droplets. Cellulose insulation was sprayed into the standard 300 x 300 mm² moulds, without glue, and laid to dry at the standard conditions. Once dry, samples were cut using a circular saw, making 90 x 90 x 300 mm^3 samples. A burner was placed at a 45° angle at the lower corner in the centre of the sample. The sample was fitted on a wire mesh holder with an opening for contact with the flame. The flame of the burner was adjusted to have a height of 20mm. According to the standard, for classifications ranging from D to B, a 30 s exposure time is required, and the burning period after the flame is removed is 60 seconds, in which the flame spread threshold of 150 mm should be maintained. A piece of filter paper is placed under the sample to test if it will ignite with the droplets from the material. The material is weighed before and after the test, and the change in mass is calculated. The smoke production from the material is observed as well. Figure E16 illustrates the setup for the test.

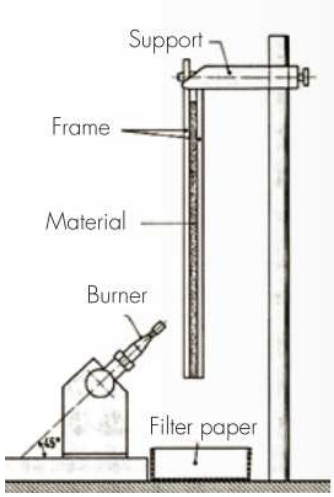


Figure E16: Flame test setup.

LIST OF FIGURES

Figure 1.1: Cellulose insulation fibres.....6

Figure 1.2: Example of a typical manufacturing process of CFI, adapted with permission from (Makron Engineering, n.d.).....8

Figure 1.3: Microscopic scan of cellulose fibres, 10X magnification.....9

Figure 1.4: Calculated minimal density for settling prevention of loose fill CFI in a wall under static humidity conditions (50% and 80% RH), as a function installed thickness, top left shows the dimensions of the wall cavity (Rasmussen, 2002) 12

Figure 1.5: Increase in thermal conductivity with moisture content of cellulose fibre insulation (Véjelis et al., 2006) 14

Figure 1.6: Proportions (parts per hundred) of borax and boric acid in order to achieve smoulder retardancy (// diagonal) and flame retardancy (\\ diagonal) (Day et al., 1980). 16

Figure 1.7: Sorption-desorption isotherm of treated cellulose insulation.....19

Figure 1.8: Calculated evolution of average moisture content in exterior facing half of wet spray CFI with varying months of installation in Detroit Michigan (Salonvaara et al., 2010).....22

Figure 1.9: Adsorption bonding adhesion (Mattson, 2010).....25

Figure 1.10: Mechanical interlocking adhesion (Mattson, 2010) 26

Figure 1.11: adsorption bonding illustration (Mattson, 2010).....26

Figure 1.12: Electrostatic adhesion (Mattson, 2010). 27

Figure 1.13: Schematic representation of microfibril formation of hydrogen bonds between cellulosic fibres (Gardner et al., 2008).....29

Figure 1.14: Structure of cellulose (Pizzi and Mittal, 2003) 31

Figure 1.15: Structure of Chitin and Chitosan (Rinaudo, 2006).....32

Figure 1.16: Partial structure of pectin (Baker et al., 2012).....33

Figure 1.17: Structure of guar gum (Mudgil et al., 2014). 34

Figure 1.18: Structure of starch components (Pizzi and Mittal, 2003).....35

Figure 1.19: Structure of three phenyl propane monomers of lignin. (Hatakeyama and Hatakeyama, 2009).....36

Figure 1.20: Structure of tannic acid. Tannin acid (Ahmad, 2014). 37

Figure 1.21: Schematic representation of types of surfactants with hydrophilic head and hydrophobic tail. 41

Figure 1.22: Water retention value vs CTAB dosage in recycled newsprint paper pulp (Beaupré (2012) 42

Figure 2.1: Loose Univercell CFI fibres.....48

Figure 2.2 : CFI fibres under X7.5 magnification.....49

Figure 2.3: CFI fibres under X20 magnification.....49

Figure 2.4: Particle size distribution of cellulose samples. 53

Figure 2.5: Types of water present in cellulosic fibre surface (Samyn, 2013). 56

Figure 2.6: Frozen and unfrozen bound water, free water and total bound water as a function of total water content for untreated Univercell CFI. 57

Figure 2.7: Dynamic vapour sorption of CFI, with Oswin model approximation. 59

Figure 2.8: Projected CFI moulds. 60

Figure 2.9: Drying of cellulose samples, comparison of different source material and treatment. 62

Figure 2.10: Drying of cellulose samples, with varying installed water pressure. 63

Figure 2.11: Influence of installed moisture on final density at equilibrium, with polynomial regression line. 64

Figure 2.12: Compression test sample. 65

Figure 2.13: Stress-strain curves of projected cellulose insulation samples with varying moisture content. 66

Figure 2.14: Influence of installed moisture content on modulus of elasticity E, with linear regression line. 67

Figure 2.15: Influence of installed moisture content on $\sigma_{5\%}$ $\sigma_{10\%,}$ with linear regression line. ... 67

Figure 2.16: Influence of final density after spraying with various water dosages on modulus of elasticity E, compared to compacted samples, with regression lines. 68

Figure 2.17: Influence of final density after spraying with various water dosages on $\sigma_{5\%}$ $\sigma_{10\%,}$ compared to compacted samples, with regression lines. 69

Figure 2.18: Influence of installed moisture content on thermal conductivity values λ_{10} and $\lambda_{25,}$ with linear regression lines. 71

Figure 2.19: Influence of dry density on thermal conductivity values λ_{10} and $\lambda_{25,}$ with linear regression lines. 72

Figure 3.1: Optimal additive formulation Venn diagram. 76

Figure 3.2: Concentration dependence of viscosity for polysaccharides, from (Sworn, 2007). .. 79

Figure 3.3: Viscosity-shear curve of pectin at varying concentration. 79

Figure 3.4: Viscosity concentration curves for binder solutions. 81

Figure 3.5: Viscosity concentration curves for shear thinning (non-Newtonian) binder solutions at high shear. 82

Figure 3.6: Tensile strength vs concentration of binder solutions. 85

Figure 3.7: Drying curves for cellulose samples. 92

Figure 3.8: final sprayable and adhesive concentrations. 92

Figure 3.9: Modulus of elasticity of samples. 96

Figure 3.10: measured stress at 5% and 10% with density of treated samples. 97

Figure 3.11: Heterogeneities of samples. 98

Figure 3.12: Cellulose-pectin samples sprayed at low dosage concentrations. 99

Figure 3.13: Thermal conductivity of cellulose insulation samples. 100

Figure 4.1: Drying cure of cellulose with lignosulfonate additives at variable concentration. . 106

Figure 4.2: Compression resistance for cellulose with lignosulfonate and CTAB additives at variable concentrations..... 107

Figure 4.3: Heterogeneity in 40% concentration lignosulfonate samples..... 108

Figure 4.4: Thermal conductivity for cellulose with lignosulfonate and CTAB additives at variable concentrations..... 109

Figure 4.4: Plotted drying times with equivalent empirical model of cellulose with varying initial dosage. 112

Figure 4.5: Influence of initial moisture on the drying time constant $t_{c..}$ 113

Figure 4.6: Plotted drying times with equivalent empirical model of cellulose with varying thickness. 114

Figure 4.7: Influence of installed thickness on the drying time constant, t_c 115

Figure 4.8: Capillary sorption measurement with sorption coefficient $A_{w.}$ 120

Figure 4.9: Moisture dependent moisture diffusivity coefficient for cellulose insulation. 121

Figure 4.10: Drying curves of cellulose with calculated and fitted capillary sorption constant $A_{w..}$ 122

Figure 4.11: WUFI wall material layers profile. 124

Figure 4.12: Modelled drying curves under different ambient and thickness conditions at an initial dosage of 40% 125

Figure 4.13: Modelled drying curves under different ambient and thickness conditions at an initial dosage of 80% 126

Figure 4.14: Time to reach dry state (20% moisture) for cellulose with varying formulations, sprayed dosage, and insulation thickness..... 128

Figure 4.15: Open flame combustion tests setup. 131

Figure 4.16: Open flame cellulose sample. 132

Figure 4.17: Mould in high moisture cellulose samples. 133

Figure 4.18: Fungal growth discoloration on cellulose samples with different additives. 134

Figure E1: Schopper Reigler freeness tester..... 146

Figure E2: Tapped density apparatus. 147

Figure E3: DVS measurement apparatus. 150

Figure E4: DSC melting curves..... 152

Figure E5: (a) Cellulose blower and wet spraying procedure (b) Wet spraying water pump (c) blowing hose with water nozzles (d). 154

Figure E6: Cellulose wet spraying..... 155

Figure E7: Excess material removal. 155

Figure E8: Compression tests. 157

Figure E9: λ-Meter EP500e apparatus. 158

Figure E10: viscosity measurement apparatus. 160

Figure E11: Tensile strength tests. 161

Figure E12: Tensiometer..... 162

Figure E13: Capillary sorption measurement setup (Hansen et al., 2001) 163
Figure E14: Water absorption coefficient determination (Hansen et al., 2001) 164
Figure E15: WUFI 5 software UI. 165
Figure E16: Flame test setup. 167

LIST OF TABLES

Table 1.1: Market share of US insulation materials in 2012 and 2013 source: (Home Innovation Research Labs, 2013) 6

Table 1.2: Average component proportions of newsprint and office paper, (Wu et al., 2001) 8

Table 1.3: Comparative life cycle analysis of common building materials (Zabalza Bribián et al., 2011) 17

Table 1.4: bond energies of primary and secondary bonds (Pizzi, 1994) 25

Table 2.1: Schopper-Reigler freeness testing 50

Table 2.2 Tapped density measurements 51

Table 2.3: Chemical composition of cellulose insulation samples 54

Table.2.4: Water retention value (WRV) of cellulose insulation samples 55

Table 2.5: Measured differences between cellulose brands and possible influences on drying. 61

Table 3.1: typical shear rates for different processes. 80

Table 3.2: Sprayable concentrations of binder solutions and viscosities. 83

Table 3.3: Sprayable concentrations of binder solutions and viscosities, at high shear rate. 83

Table 3.4: Comparison of tensile strength of binder solutions. 86

Table 3.5: Comparison of tensile strength of non-Newtonian binder solutions with high shear. 86

Table 3.6: Final sprayable and adhesive concentrations. 87

Table 3.7: Pump rate of additives. 89

Table 3.8: Properties of surfactants. 90

Table 3.9: Surface tension measurements. 90

Table 3.9: Drying times of surfactant samples. 94

Table 4.1: Time to reach dry state for cellulose with lignosulfonate additives at variable concentration. 106

Table 4.2: Indicative drying times for cellulose insulation as a function of thickness and ambient conditions (CSTB, 2011) 110

Table 4.3: Constants for empirical model for drying of cellulose with initial moisture dosage. 112

Table 4.4: Constants for empirical model for additive formulations 115

Table 4.5: Specific heat capacity of cellulose insulation formulations. 118

Table 4.6: Measured capillary sorption coefficient $A_{w.}$ 121

Table 4.7: Values of fitted capillary sorption constant A_w 123

Table 4.8: Relative decrease in drying times via the contribution of lignosulfonate additives at 28% water concentration. 128

Table 4.9: Smouldering combustion test results. 130

Table 4.10: Open flame combustion tests. 131

Table 4.11: Properties summary of cellulose insulation with lignosulfonate and CTAB additives. 135

Table 4.12: Potential impact of additives on primary energy demand and global warming potential of cellulose insulation. 137

Table E01: Experimental run design parameters. 166

BIBLIOGRAPHY

AFNOR, 2014. NF EN 15101-1: Thermal insulation products for buildings — In-situ formed loose fill cellulose (LFCI) products — Part 1: Specification for the products before installation.

Ahmad, T., 2014. Reviewing the Tannic Acid Mediated Synthesis of Metal Nanoparticles. Journal of Nanotechnology 2014, 1–11. doi:10.1155/2014/954206

Alibaba.com, 2016. Material price search, Ali Baba [WWW Document]. URL https://french.alibaba.com (accessed 5.26.16).

Alila, S., Boufi, S., Belgacem, M.N., Beneventi, D., 2005. Adsorption of a Cationic Surfactant onto Cellulosic Fibers I. Surface Charge Effects. Langmuir 21, 8106–8113. doi:10.1021/la050367n

Alvarez, F.X., Jou, D., Sellitto, A., 2010. Pore-size dependence of the thermal conductivity of porous silicon: A phonon hydrodynamic approach. Applied Physics Letters 97, 33103. doi:10.1063/1.3462936

Amziane, S., Arnaud, L., 2013. Les bétons de granulats d'origine végétale : Application au béton de chanvre. Lavoisier.

ASTM, 2006. ASTM C739 Standard Specification for Cellulosic Fiber Loose File Thermal Insulation.

Baker, C.G.J., Ranken, M.D., Kill, R.C., 2012. Food Industries Manual. Springer Science & Business Media.

Barreira, E., Delgado, J.M., de Freitas, V.P., 2014. Wetting and Drying Kinetics of Building Materials, in: Drying and Wetting of Building Materials and Components. Springer, pp. 51–69.

Baumann, M.G., Conner, A.H., 1994. Carbohydrate polymers as adhesives. Handbook of adhesive technology 495–510.

Beaupré, J.J., 2012. Chemically enhanced water removal in papermaking. University of Maine.

Benitha Sandrine, U., Isabelle, V., Ton Hoang, M., Chadi, M., 2015. Influence of chemical modification on hemp–starch concrete. Construction and Building Materials 81, 208–215. doi:10.1016/j.conbuildmat.2015.02.045

Biermann, C.J., 1993. Essentials of pulping and papermaking. Academic Press.

Binder, A., Zirkelbach, D., Künzel, H., 2007. New method to quantify liquid transport inside hydrophilic interior insulation materials.

Bisanda, E.T.N., Ogola, W.O., Tesha, J.V., 2003. Characterisation of tannin resin blends for particle board applications. Cement and Concrete Composites 25, 593–598.

Bomberg, M., Shirtliffe, C.J., 1979. Comments on Standardization of Density and Thermal Resistance Testing of Cellulose Fibre Insulation for Horizontal Applications. Building Research Note 147, 17.

Bomberg, M., Solvason, K.R., 1980. How To Ensure Good Thermal Performance of Cellulose Fiber Insulation. Part II. Exterior Walls. Journal of Building Physics 4, 119–133. doi:10.1177/109719638000400204

Boogman, D.P., 2014. EPD - ENVIRONMENTAL PRODUCT DECLARATION Blown insulation made of cellulose fibre ISOCELL GmbH.

Boonyakarn, S., D., A., Spiezle, S.R., 1990. Fibreglass vs cellulose installed performance.
Brancato, A.A., 2008. Effect of progressive recycling on cellulose fiber surface properties. ProQuest.
British Standards Institute, 2002. BS EN ISO 15148:2002 Hygrothermal performance of building materials and products, Determination of water absorption coefficient by partial immersion,.
Brother, G.H., Smith, A.K., 1940. Aqueous dispension of hardened protein. US2210481 A.
Bye, C.N., 1990. Casein and mixed protein adhesives, in: Handbook of Adhesives. Springer, pp. 135–152.
Cerolini, S., D'Orazio, M., Di Perna, C., Stazi, A., 2009. Moisture buffering capacity of highly absorbing materials. Energy and Buildings 41, 164–168. doi:10.1016/j.enbuild.2008.08.006
CIMA, 1998a. CIMA Technical Bulletin #2 Standard Practice for Installing Cellulose Building Insulation.
CIMA, 1998b. CIMA Technical Bulletin #3 Standard Practice for Installation of Sprayed Cellulosic Wall Cavity Insulation.
CMHC, 1990. Wet-sprayed Cellulose Insulation in wood frame construction, Research & Development Highlights Technical Series 90-240. Edmonton Canada.
Conner, A.H., 1990. Adhesives from renewable resources: ACS Symposium Series No. 385 Edited by R. W. Hemingway and A. H. Conner, American Chemical Society, Washington, 1989. pp. ix + 510, price $99.95 (US/Canada)/$119.95 (Export). ISBN 0–8412–1562–6. Brit. Poly. J. 23, 185–186. doi:10.1002/pi.4980230132
Coppens, H.A., Santana, M.A.E., Pastore, F.J., others, 1980. Tannin formaldehyde adhesive for exterior-grade plywood and particleboard manufacture. Forest Products Journal 30, 38–42.
CSTB, 2011. Technical approval "Univercell Confort Vrac" for dense pack and wet-spray installation (in French).
Czerwin, E.P., 1978. Process of preparing corrugated paper board with a particular polyvinyl alcohol modified starch-based corrugating adhesive. Google Patents.
Day, M., Suprunchuk, T., Wiles, D.M., 1981. The Fire Properties of Cellulose Insulation. Journal of Building Physics 4, 157–170. doi:10.1177/109719638100400301
Day, M., Suprunchuk, T., Wiles, D.M., 1980. A Combustibility Study of Cellulose Insulation. Journal of Building Physics 3, 260–271. doi:10.1177/109719638000300404
Day, M., Wiles, D.M., 1978. Combustibility of Loose Fiber Fill Cellulose Insulation: the Role of Borax and Boric Acid. Journal of Building Physics 2, 30–39. doi:10.1177/109719637800200104
Delgado, J., Ramos, N.M.M., De Freitas, V.P., 2006. Can moisture buffer performance be estimated from sorption kinetics? Journal of building physics 29, 281–299.
Diniz, J.F., Gil, M.H., Castro, J., 2004. Hornification—its origin and interpretation in wood pulps. Wood Science and Technology 37, 489–494.
Dragostin, O.M., Samal, S.K., Dash, M., Lupascu, F., Pânzariu, A., Tuchilus, C., Ghetu, N., Danciu, M., Dubruel, P., Pieptu, D., Vasile, C., Tatia, R., Profire, L., 2016. New antimicrobial chitosan derivatives for wound dressing applications. Carbohydrate Polymers 141, 28–40. doi:10.1016/j.carbpol.2015.12.078

Ebnesajjad, S. (Ed.), 2008. Adhesives technology handbook, 2nd ed. ed. William Andrew Pub, Norwich, NY.
Elfordy, S., Lucas, F., Tancret, F., Scudeller, Y., Goudet, L., 2008. Mechanical and thermal properties of lime and hemp concrete ("hempcrete") manufactured by a projection process. Construction and Building Materials 22, 2116–2123. doi:10.1016/j.conbuildmat.2007.07.016
Eloukabi, H., Sghaier, N., Ben Nasrallah, S., Prat, M., 2013. Experimental study of the effect of sodium chloride on drying of porous media: The crusty–patchy efflorescence transition. International Journal of Heat and Mass Transfer 56, 80–93. doi:10.1016/j.ijheatmasstransfer.2012.09.045
EN, 2010. 13501-1+ A1: 2010 Fire classification of construction products and building elements. Part.
EN, B., 2002. 13823: 2002. Reaction to fire tests for building products: building products excluding floorings exposed to the thermal attack by a single burning item. British Standards Institution, London, UK.
Eriksson, M., 2006. The influence of molecular adhesion on paper strength.
European Parliament, 2010. DIRECTIVE 2010/31/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 19 May 2010 on the energy performance of buildings (recast).
Fischer Jr, A.C., Mcelmury, D.E., 1969. Single ungelatinized starch-component-corrugating adhesive. Google Patents.
Flory, A.R., Vicuna Requesens, D., Devaiah, S.P., Teoh, K.T., Mansfield, S.D., Hood, E.E., 2013. Development of a green binder system for paper products. BMC Biotechnology 13, 28. doi:10.1186/1472-6750-13-28
Floyd, W.C., Thompson, N., Dragner, L.R., 2001. Manufacturing paper with improved strength by introducing into the paper pulp in the wet end of a paper making process an aqueous starch dispersion of a gelatinized cationic starch and a blocked glyoxal resin. Google Patents.
Fornué, E.D., Allan, G.G., Quiñones, H.J.C., González, G.T., Saucedo, J.T., 2011. Fundamental aspects of adhesion between cellulosic surfaces in contact—a review. O Papel 72, 85–90.
Fourche, G., 1995. An overview of the basic aspects of polymer adhesion. Part I: Fundamentals. Polymer Engineering and Science 35, 957–967. doi:10.1002/pen.760351202
Fraga, I., Montserrat, S., Hutchinson, J.M., 2007. TOPEM, a new temperature modulated DSC technique. Journal of thermal analysis and calorimetry 87, 119–124.
Frihart, C., 2010. Part 2 Biobased Adhesives And Non-Convential Bonding. Series: Journal Articles.
Gardner, D.J., Oporto, G.S., Mills, R., Samir, M.A.S.A., 2008. Adhesion and Surface Issues in Cellulose and Nanocellulose. Journal of Adhesion Science and Technology 22, 545–567. doi:10.1163/156856108X295509
Gharehkhani, S., Sadeghinezhad, E., Kazi, S.N., Yarmand, H., Badarudin, A., Safaei, M.R., Zubir, M.N.M., 2015. Basic effects of pulp refining on fiber properties—A review. Carbohydrate Polymers 115, 785–803. doi:10.1016/j.carbpol.2014.08.047
Godish, T.J., Godish, D.R., 2006. Mold infestation of wet spray-applied cellulose insulation. J Air Waste Manag Assoc 56, 90–95.

González-García, S., Hospido, A., Agnemo, R., Svensson, P., Selling, E., Moreira, M.T., Feijoo, G., 2011. Environmental Life Cycle Assessment of a Swedish Dissolving Pulp Mill Integrated Biorefinery. Journal of Industrial Ecology 15, 568–583. doi:10.1111/j.1530-9290.2011.00354.x
Gösta, B., 2003. Dairy Processing Handbook, 1 Edition edition. ed. Tetra Pak Processing Systems AB, Lund, Sweden.
Graves, R.S., Yarbrough, D.W., 1989. An experimental study of stabilized cellulosic insulation installed in four attic sections of manufactured homes.
Grossi, F.X., 1954. Wallpaper paste.
Guo, J.-H., Skinner, G.., Harcum, W.., Barnum, P.., 1998. Pharmaceutical applications of naturally occurring water-soluble polymers. Pharmaceutical Science & Technology Today 1, 254–261. doi:10.1016/S1461-5347(98)00072-8
Haars, A., Huttermann, A., 1984. Process for producing a binder for wood materials. Google Patents.
Hagentoft, C.-E., Harderup, E., 1996. Moisture Conditions in a North Facing Wall with Cellulose Loose Fill Insulation: Constructions with and without Vapor Retarder and Air Leakage. Journal of Building Physics 19, 228–243. doi:10.1177/109719639601900304
Hansen, K.K., Rode, C., Hansen, E.J. de P., Padfield, T., Kristiansen, F.H., 2001. Experimental investigation of the hygrothermal performance of insulation materials. Performance of exterior envelopes of whole buildings VIII.
Hatakeyama, H., Hatakeyama, T., 2009. Lignin Structure, Properties, and Applications, in: Abe, A., Dusek, K., Kobayashi, S. (Eds.), Biopolymers, Advances in Polymer Science. Springer Berlin Heidelberg, pp. 1–63.
Henry, Bradshaw, L., 1931. Process of making slow-setting casein glue, and dry base for use in such processes. US1829259 A.
Herrera, J., 2005. Assessment of Fungal Growth on Sodium Polyborate-Treated Cellulose Insulation. Journal of Occupational and Environmental Hygiene 2, 626–632. doi:10.1080/15459620500377667
Hilado, C.J., Olcomendy, E.M., Brauer, D.P., 1979. Toxicity of Gases Generated From Smoldering Cellulose Insulation. Journal of Building Physics 2, 131–134. doi:10.1177/109719637900200302
Home Innovation Research Labs, 2013. Annual Builder Practices Survey, 2013.
Huang, H., 2008. Life cycle assessment comparison between Pepfactant® and chemical surfactant production.
Hubbe, M.A., Venditti, R.A., Rojas, O.J., 2007. What happens to cellulosic fibres during papermaking and recycling? A review. BioResources; Vol 2, No 4 (2007).
Hussein, A.S., Ibrahim, K.I., Abdulla, K.M., 2011. Tannin–Phenol Formaldehyde Resins As Binders for Cellulosic Fibers: Mechanical Properties. Natural Resources 2, 98–101. doi:10.4236/nr.2011.22013
Imam, S.H., Bilbao-Sainz, C., Chiou, B.-S., Glenn, G.M., Orts, W.J., 2013. Biobased adhesives, gums, emulsions, and binders: current trends and future prospects. Journal of Adhesion Science and Technology 27, 1972–1997. doi:10.1080/01694243.2012.696892
Imam, S.H., Gordon, S.H., Mao, L., Chen, L., 2001. Environmentally friendly wood adhesive from a renewable plant polymer: characteristics and optimization.

Polymer Degradation and Stability 73, 529–533. doi:10.1016/S0141-3910(01)00114-8
Imam, S.H., Mao, L., Chen, L., Greene, R.V., 1999. Wood Adhesive from Crosslinked Poly(Vinyl Alcohol) and Partially Gelatinized Starch: Preparation and Properties. Starch/Stärke 51, 225–229. doi:10.1002/(SICI)1521-379X(199906)51:6<225::AID-STAR225>3.0.CO;2-F
ISO, E., 2002. 11925-2: 2002. Reaction to fire tests-Ignitability of building products subjected to direct impingement of flame-Part 2.
Johnson, L.A., Myers, D.J., Burden, D.J., others, 1984. Early uses of soy protein in far east. Inform 3, 282–284.
Kalamees, T., Vinha, J., 2003. Hygrothermal calculations and laboratory tests on timber-framed wall structures. Building and Environment 38, 689–697. doi:10.1016/S0360-1323(02)00207-X
Karagiozis, A., Künzel, H.M., Holm, A., 2001. WUFI ORNL/IBP–A North American Hygrothermal Model: Proceedings of the Thermal Performance of the Exterior Envelopes of Whole Buildings VIII (Buildings VIII). Clearwater Beach, Florida, USA.
Kazayawoko, J.-S., Riedl, B., Poliquin, J., Barry, A.O., Matuana, L.M., 1992. A lignin-phenol-formaldehyde binder for particleboard Part 1. Thermal characteristics. Holzforschung-International Journal of the Biology, Chemistry, Physics and Technology of Wood 46, 257–262.
Kendall, K., 1994. Adhesion: Molecules and Mechanics. Science 263, 1720–1725. doi:10.1126/science.263.5154.1720
Khosravi, S., Nordqvist, P., Khabbaz, F., Johansson, M., 2011. Protein-based adhesives for particleboards—Effect of application process. Industrial Crops and Products 34, 1509–1515. doi:10.1016/j.indcrop.2011.05.009
Khosravi, S., Nordqvist, P., Khabbaz, F., Öhman, C., Bjurhager, I., Johansson, M., 2015. Wetting and film formation of wheat gluten dispersions applied to wood substrates as particle board adhesives. European Polymer Journal 67, 476–482. doi:10.1016/j.eurpolymj.2014.11.034
Kim, S., 2009. Environment-friendly adhesives for surface bonding of wood-based flooring using natural tannin to reduce formaldehyde and TVOC emission. Bioresource Technology 100, 744–748. doi:10.1016/j.biortech.2008.06.062
Kim, S., Lee, Y.-K., Kim, H.-J., Lee, H.H., 2003. Physico-mechanical properties of particleboards bonded with pine and wattle tannin-based adhesives. Journal of Adhesion Science and Technology 17, 1863–1875. doi:10.1163/156856103770572025
Klašnja, B., Kopitović, S., 1992. Lignin-Phenol-Formaldehyde resins as adhesives in the production of plywood. Holz als Roh- und Werkstoff 50, 282–285. doi:10.1007/BF02615352
Krinski, T.L., Coco, C.E., Steinmetz, A.L., 1987. Modified protein adhesive binder and process for producing using cationic monomers. US4689381 A.
Krinski, T.L., Tran, T.H., Gambaro, J.J., 1990. Adhesive binders for paper coating composition having improved stability and whiteness. US4961788 A.
Krus, M., Holm, A., 1999. Simple methods to approximate the liquid transport coefficients describing the absorption and drying process, in: Proceedings of the

5th Symposium'Building Physics in the Nordic Countries', Göteborg. pp. 241–248.
Kumar, R., Choudhary, V., Mishra, S., Varma, I.K., Mattiason, B., 2002. Adhesives and plastics based on soy protein products. Industrial Crops and Products 16, 155–172. doi:10.1016/S0926-6690(02)00007-9
Künzel, H.M., 1995. Simultaneous heat and moisture transport in building components. One-and two-dimensional calculation using simple parameters. IRB-Verlag Stuttgart.
Kuo, M., Adams, D., Myers, D., Curry, D., others, 1998. Properties of wood/agricultural fiberboard bonded with soybean-based adhesives. Forest Products Journal 48, 71.
Kwon, Y.C., Yarbrough, D.W., 2004. A Comparison of Korean Cellulose Insulation with Cellulose Insulation Manufactured in the United States of America. Journal of Thermal Envelope and Building Science 27, 185–197. doi:10.1177/1097196304035242
Lacasta Palacio, A.M., Palumbo Fernández, M., Navarro Ezquerra, A., Avellaneda Díaz-Grande, J., others, 2014. Characterization of thermal insulation materials developed with crop wastes and natural binders.
Lambuth, A.L., 2003. Protein adhesives for wood. Handbook of adhesive technology 2.
Le, A.T., Gacoin, A., Li, A., Mai, T.H., Rebay, M., Delmas, Y., 2014. Experimental investigation on the mechanical performance of starch–hemp composite materials. Construction and Building Materials 61, 106–113.
Lin, H., Gunasekaran, S., 2010. Cow blood adhesive: Characterization of physicochemical and adhesion properties. International Journal of Adhesion and Adhesives 30, 139–144. doi:10.1016/j.ijadhadh.2009.10.003
Lorenz, L.F., Conner, A.H., Christiansen, A.W., 1999. Effect of soy protein additions on the reactivity and formaldehyde emissions of urea-formaldehyde adhesive resins. Forest products journal 49, 73–78.
Madad, N., Chebil, L., Sanchez, C., Ghoul, M., 2011. Effect of molecular weight distribution on chemical, structural and physicochemical properties of sodium lignosulfonates. RASAYAN J Chem 4, 189–202.
Makron Engineering, n.d. Fibretec 1500 production line.
Mancera, C., El Mansouri, N.-E., Vilaseca Morera, F., Ferrando, F., Salvado, J., 2011. The effect of lignin as a natural adhesive on the physico-mechanical properties of Vitis vinifera fiberboards. \copyright BioResources, 2011, vol. 6, núm. 3, p. 2851-2860.
Manfred, K., Schmidt, T., 2008. Radiation effects on exterior surfaces, in: Proceedings of the 8th Symposium on Building Physics in Nordic Countries. Technical University of Denmark, Copenhagen, Denmark, pp. 207–212.
Marchand, R.G., Kumaran, M.K., 1994. Moisture Diffusivity of Cellulose Insulation. Journal of Building Physics 17, 362–377. doi:10.1177/109719639401700406
Mati-Baouche, N., De Baynast, H., Lebert, A., Sun, S., Lopez-Mingo, C.J.S., Leclaire, P., Michaud, P., 2014a. Mechanical, thermal and acoustical characterizations of an insulating bio-based composite made from sunflower stalks particles and chitosan. Industrial Crops and Products 58, 244–250. doi:10.1016/j.indcrop.2014.04.022
Mati-Baouche, N., Elchinger, P.-H., de Baynast, H., Pierre, G., Delattre, C., Michaud, P., 2014b. Chitosan as an adhesive. European Polymer Journal 60, 198–212. doi:10.1016/j.eurpolymj.2014.09.008

Mattson, C., 2010. Physics of adhesives.
Menaker, A.J., 1938. Method of preparing a casein paint. Google Patents.
Mittal, M., Sharma, C.B., 1992. Studies on lignin-based adhesives for plywood panels. Polymer International 29, 7–8. doi:10.1002/pi.4990290103
Morgan, D.L., 2006. NTP Toxicity Study Report on the atmospheric characterization, particle size, chemical composition, and workplace exposure assessment of cellulose insulation (CELLULOSEINS). Toxic Rep Ser 1–62, A1-C2.
Mortensen, L.H., Peuhkuri, R., Rode, C., 2005. Full scale tests of moisture buffer capacity of wall materials, in: 7 Th Nordic Symposium on Building Physics, Reykjavík. pp. 662–669.
Mudgil, D., Barak, S., Khatkar, B.S., 2014. Guar gum: processing, properties and food applications—A Review. Journal of Food Science and Technology 51, 409–418. doi:10.1007/s13197-011-0522-x
NAHB Research Center, 2009. Air Infiltration of Wood Frame Walls.
Nakamura, K., Hatakeyama, T., Hatakeyama, H., 1981. Studies on bound water of cellulose by differential scanning calorimetry. Textile research journal 51, 607–613.
Nasatto, P., Pignon, F., Silveira, J., Duarte, M., Noseda, M., Rinaudo, M., 2015. Methylcellulose, a Cellulose Derivative with Original Physical Properties and Extended Applications. Polymers 7, 777–803. doi:10.3390/polym7050777
Nicholson, C., Abercrombie, J., Botterill, W., Brocato, R., Detzel, J., Fitzpatrick Jr, J., Martin, C., Messer, M., Mohr, F., Robins, B., 1991. History of adhesives. ESC Reports 1.
Nicolajsen, A., 2005. Thermal transmittance of a cellulose loose-fill insulation material. Building and Environment 40, 907–914. doi:10.1016/j.buildenv.2004.08.025
Norström, E., Fogelström, L., Nordqvist, P., Khabbaz, F., Malmström, E., 2014. Gum dispersions as environmentally friendly wood adhesives. Industrial Crops and Products 52, 736–744. doi:10.1016/j.indcrop.2013.12.001
Nunthanid, J., Laungtana-anan, M., Sriamornsak, P., Limmatvapirat, S., Puttipipatkhachorn, S., Lim, L.Y., Khor, E., 2004. Characterization of chitosan acetate as a binder for sustained release tablets. Journal of Controlled Release 99, 15–26. doi:10.1016/j.jconrel.2004.06.008
Pachauri, R.K., Allen, M.R., Barros, V.R., Broome, J., Cramer, W., Christ, R., Church, J.A., Clarke, L., Dahe, Q., Dasgupta, P., 2014. Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change.
Patel, A.K., Michaud, P., de Baynast, H., Grédiac, M., Mathias, J.-D., 2013. Preparation of chitosan-based adhesives and assessment of their mechanical properties. J. Appl. Polym. Sci. 127, 3869–3876. doi:10.1002/app.37685
Pearson, C.L., 2003. Animal glues and adhesives. Handbook of adhesive technology 479–494.
Pel, L., Brocken, H., Kopinga, K., 1996. Determiination of moisture diffusivity in porous media using moisture concentration profiles. International Journal of Heat and Mass Transfer 39, 1273–1280.
Peshkova, S., Li, K., 2003. Investigation of chitosan–phenolics systems as wood adhesives. Journal of Biotechnology 102, 199–207. doi:10.1016/S0168-1656(03)00026-9

Peuhkuri, R., Rode, C., Hansen, K.K., 2008. Non-isothermal moisture transport through insulation materials. Building and Environment 43, 811–822. doi:10.1016/j.buildenv.2007.01.021
Pizzi, A., 1994. Advanced wood adhesives technology. M. Dekker, New York.
Pizzi, A., 1982. Pine tannin adhesives for particleboard. Holz als Roh-und Werkstoff 40, 293–301.
Pizzi, A., Merlin, M., 1981. A new class of tannin adhesives for exterior particleboard. International Journal of Adhesion and Adhesives 1, 261–264.
Pizzi, A., Mittal, K.L., 2003. Handbook of adhesive technology, revised and expanded. CRC Press.
Raeker, M., Johnson, L.A., 1995. Thermal and functional properties of bovine blood plasma and egg white proteins. Journal of Food Science 60, 685–690.
Rasmussen, T.V., 2005. Prediction of Density for Prevention of Settling of Hygroscopic and Nonhygroscopic Loose-fill Insulation in Walls. Journal of Thermal Envelope and Building Science 28, 245–267. doi:10.1177/1097196305048596
Rasmussen, T.V., 2003. Verification of Density Predicted for Prevention of Settling of Loose-fill Cellulose Insulation in Walls. Journal of Thermal Envelope and Building Science 27, 49–69. doi:10.1177/1097196303027001004
Rasmussen, T.V., 2002. Modelling Settling of Loose-fill Insulation in Walls, Part II Determination of Coefficients. Journal of Building Physics 25, 189–208. doi:10.1177/0075424202025003605
Rasmussen, T.V., 2001. Modelling Settling of Loose-Fill Insulation in Walls. Part I. Journal of Thermal Envelope and Building Science 25, 129–141. doi:10.1106/F5UH-5A2X-9VRA-FHPF
Rifai, M.M., El Hadidi, N.M., 2010. Investigation and analysis of three gilded wood samples from the tomb of Tutankhamun. Dawson, J., Rozeik, C and Wright, MM; Decorated Surfaces on Ancient Egyptian Objects, Technology, Deterioration and Conservation 16–24.
Rinaudo, M., 2006. Chitin and chitosan: Properties and applications. Progress in Polymer Science 31, 603–632. doi:10.1016/j.progpolymsci.2006.06.001
Rode, C., 1998. Organic Insulation Materials, the Effect on Indoor Humidity, and the Necessity of a Vapor Barrier. Proceedings, Thermal Performance of the Exterior Envelope of Buildings VII 109–121.
Rode, C., Peuhkuri, R., Time, B., Svennberg, K., Ojanen, T., Mukhopadhyaya, P., Kumaran, M., Dean, S.W., 2007. Moisture Buffer Value of Building Materials. Journal of ASTM International 4, 100369. doi:10.1520/JAI100369
Roffael, E., Dix, B., Okum, J., 2000. Use of spruce tannin as a binder in particleboards and medium density fiberboards (MDF). European Journal of Wood and Wood Products 58, 301–305.
Salonvaara, M., Pazera, M., Karagiozis, A., 2010. Impact of Weather on Predicting Drying Characteristics of Spray-Applied Cellulose Insulation.
Samyn, P., 2013. Wetting and hydrophobic modification of cellulose surfaces for paper applications. Journal of Materials Science 48, 6455–6498.
Sandberg, P.I., 1992. Determination of the effects of moisture content on the thermal transmisivity of cellulose fiber loose-fill Insulation. Thermal Performance of the Exterior Envelopes of Building III, ASHRAE/DOE/BTECC/ CIBSE conference 517–25.

Schellmann, N.C., 2007. Animal glues: a review of their key properties relevant to conservation. Studies in Conservation 52, 55–66.
Schiavoni, S., D′Alessandro, F., Bianchi, F., Asdrubali, F., 2016. Insulation materials for the building sector: A review and comparative analysis. Renewable and Sustainable Energy Reviews 62, 988–1011. doi:10.1016/j.rser.2016.05.045
Schmidt, A.C., Jensen, A.A., Clausen, A.U., Kamstrup, O., Postlethwaite, D., 2004. A comparative life cycle assessment of building insulation products made of stone wool, paper wool and flax. The International Journal of Life Cycle Assessment 9, 53–66.
Schnitzler, S., 2006. Autoclaved Aerated Concrete as a Green Building Material. Applied Research Paper.
Scholz, H.A., 1953. History of water-thinned paints. Industrial & Engineering Chemistry 45, 709–711.
Selbo, M.L., others, 1960. Adhesives, their use and performance in structural lumber production. Report. United States Forest Products Laboratory.
Sharma, B.R., Naresh, L., Dhuldhoya, N.C., Merchant, S.U., Merchant, U.C., 2006. An overview on pectins. Times Food Processing Journal 23, 44–51.
Siddiqui, S.A., 1989. Handbook on Cellulose Insulation.
Sprague, R.W., 1979. Present Variability in Chemical Add-on in the Cellulose Insulation Industry. Journal of Building Physics 2, 197–204. doi:10.1177/109719637900200403
Sprague, R.W., Shen, K.K., 1979. The Use of Boron Products in Cellulose Insulation. Journal of Building Physics 2, 161–174. doi:10.1177/109719637900200401
Stavins, R., Ji, Z., Brewer, T., Conte Grand, M., den Elzen, M., Finus, M., Gupta, J., Höhne, N., Lee, M.K., Michaelowa, A., 2014. International cooperation: Agreements and instruments. Climate Change 2014: Mitigation of Climate Change, Working Group III contribution to the IPCC 5th Assessment Report.
Svennerstedt, B., 1995. Analytical Models for Settling of Attic Loose-Fill Thermal Insulation. Journal of Building Physics 19, 189–201. doi:10.1177/109719639501900208
Sworn, G., 2007. Natural Thickeners, in: Williams, P.A. (Ed.), Handbook of Industrial Water Soluble Polymers. Blackwell Publishing Ltd, pp. 10–31.
Takano, A., Hughes, M., Winter, S., 2014. A multidisciplinary approach to sustainable building material selection: A case study in a Finnish context. Building and Environment 82, 526–535. doi:10.1016/j.buildenv.2014.09.026
Talukdar, P., Osanyintola, O.F., Olutimayin, S.O., Simonson, C.J., 2007. An experimental data set for benchmarking 1-D, transient heat and moisture transfer models of hygroscopic building materials. Part II: Experimental, numerical and analytical data. International Journal of Heat and Mass Transfer 50, 4915–4926. doi:10.1016/j.ijheatmasstransfer.2007.03.025
Tettey, U.Y.A., Dodoo, A., Gustavsson, L., 2014. Effects of different insulation materials on primary energy and CO2 emission of a multi-storey residential building. Energy and Buildings 82, 369–377. doi:10.1016/j.enbuild.2014.07.009
Thakur, B.R., Singh, R.K., Handa, A.K., Rao, M.A., 1997. Chemistry and uses of pectin — A review. Critical Reviews in Food Science and Nutrition 37, 47–73. doi:10.1080/10408399709527767

Tye, R.P., Spinney, S.C., 1979. A Study of the Effects of Moisture Vapour on the Thermal Transmittance Characteristics of Cellulose Fibre Thermal Insulation. Journal of Building Physics 2, 175–196. doi:10.1177/109719637900200402
UNEP, 2007. Buildings and climate change: status, challenges, and opportunities. UNEP DTIE, Sustainable Consumption and Production Branch, Nairobi, Kenya : United Nations Development Programme ; Paris, France.
Van Soest, P.J., Wine, R.H., others, 1968. Determination of lignin and cellulose in acid-detergent fiber with permanganate. Journal of the association of official analytical chemists 51, 780–785.
Van Soest, P. und, Wine, R.H., 1967. Use of detergents in the analysis of fibrous feeds. IV. Determination of plant cell-wall constituents. J. Assoc. Off. Anal. Chem 50, 50–55.
Vaughn, S.F., Kenar, J.A., Felker, F.C., Berhow, M.A., Cermak, S.C., Evangelista, R.L., Fanta, G.F., Behle, R.W., Lee, E., 2013. Evaluation of alternatives to guar gum as tackifiers for hydromulch and as clumping agents for biodegradable cat litter. Industrial Crops and Products 43, 798–801. doi:10.1016/j.indcrop.2012.09.002
Véjelis, S., Gnipas, I., Kersulis, V., 2006. Performance of Loose-Fill Cellulose Insulation. Materials Science (medziagotyra).
Vieira, D.B., Carmona-Ribeiro, A.M., 2006. Cationic lipids and surfactants as antifungal agents: mode of action. J. Antimicrob. Chemother. 58, 760–767. doi:10.1093/jac/dkl312
Vo Hong, N., Pyka, G., Wevers, M., Goderis, B., Van Puyvelde, P., Verpoest, I., Van Vuure, A.W., 2015. Processing rigid wheat gluten biocomposites for high mechanical performance. Composites Part A: Applied Science and Manufacturing 79, 74–81. doi:10.1016/j.compositesa.2015.09.006
Voiutskii, S.S., 1963. Autohesion and adhesion of high polymers.
Vrána, T., Gudmundsson, K., 2010. Comparison of fibrous insulations – Cellulose and stone wool in terms of moisture properties resulting from condensation and ice formation. Construction and Building Materials 24, 1151–1157. doi:10.1016/j.conbuildmat.2009.12.026
Wagle, D.G., Yasnovsky, V.M., 1990. Heat treatment of paper products having starch additives. Google Patents.
Wang, D., Sun, X.S., 2002. Low density particleboard from wheat straw and corn pith. Industrial Crops and Products 15, 43–50. doi:10.1016/S0926-6690(01)00094-2
Wang, Z., Li, Z., Gu, Z., Hong, Y., Cheng, L., 2012. Preparation, characterization and properties of starch-based wood adhesive. Carbohydrate Polymers 88, 699–706. doi:10.1016/j.carbpol.2012.01.023
Weise, U., Maloney, T., Paulapuro, H., 1996. Quantification of water in different states of interaction with wood pulp fibres. Cellulose 3, 189–202. doi:10.1007/BF02228801
Widsten, P., Kandelbauer, A., 2014. Industrial scale evaluation of cationic tannin as a binder for hardboard. Journal of Adhesion Science and Technology 28, 1256–1263. doi:10.1080/01694243.2014.896068
Wistara, N., Young, R.A., 1999. Properties and treatments of pulps from recycled paper. Part I. Physical and chemical properties of pulps. Cellulose 6, 291–324. doi:10.1023/A:1009221125962

Wu, B., Taylor, C.M., Knappe, D.R.U., Nanny, M.A., Barlaz, M.A., 2001. Factors Controlling Alkylbenzene Sorption to Municipal Solid Waste. Environ. Sci. Technol. 35, 4569–4576. doi:10.1021/es010893a

Wu, Y.B., Lv, C.F., Han, M.N., 2009. Synthesis and Performance Study of Polybasic Starch Graft Copolymerization Function Materials. Advanced Materials Research 79–82, 43–46. doi:10.4028/www.scientific.net/AMR.79-82.43

Yan, D., Li, K., 2012. Evaluation of inter-fiber bonding in wood pulp fibers by chemical force microscopy. Journal of Materials Science Research 2, p23.

Zabalza Bribián, I., Valero Capilla, A., Aranda Usón, A., 2011. Life cycle assessment of building materials: Comparative analysis of energy and environmental impacts and evaluation of the eco-efficiency improvement potential. Building and Environment 46, 1133–1140. doi:10.1016/j.buildenv.2010.12.002

Zaborniak, R., 1989. Prediction of Long-Term Density of Cellulose Fibre Insulation in Horizontal Spaces of New Residential Houses. Journal of Building Physics 13, 21–32. doi:10.1177/109719638901300104

Zhang, Y., Ding, L., Gu, J., Tan, H., Zhu, L., 2015. Preparation and properties of a starch-based wood adhesive with high bonding strength and water resistance. Carbohydrate Polymers 115, 32–37. doi:10.1016/j.carbpol.2014.08.063

Zhong, Z., Sun, X.S., Fang, X., Ratto, J.A., 2001. Adhesion properties of soy protein with fiber cardboard. J Amer Oil Chem Soc 78, 37–41. doi:10.1007/s11746-001-0216-0

RESUME EN FRANÇAIS

Introduction

Les matériaux d'isolation sont une partie essentielle de l'enveloppe du bâtiment. Ils assurent que la température à l'intérieur d'une chambre est maintenue à un certain niveau, et à réduire l'utilisation de chauffage et de refroidissement, qui ont une forte demande d'énergie. Actuellement, la majorité des matériaux isolants traditionnels tels que la laine minérale sont fabriqués à partir de ressources non renouvelables.

Un matériau d'isolation alternatif, la ouate de cellulose, a l'avantage d'être fabriqué à partir du papier recyclé, tout en ayant des propriétés de performance similaires aux matériaux d'isolation traditionnels. Habituellement, l'isolant cellulosique est soufflé à sec dans les cavités murales fermées ou les toitures. Une méthode plus récente de l'installation de la ouate de cellulose, la voie humide, présente les avantages d'assurer une bonne distribution et le remplissage de la matière dans une cavité de mur, tout en empêchant le tassement grâce à l'utilisation de l'eau pulvérisée. Les inconvénients associés à cette méthode, cependant, sont principalement dues à une mauvaise installation de la ouate de cellulose alors qu'il est pulvérisé avec de l'eau. Si trop d'eau est appliqué, ou si son application dans une humidité élevée, des conditions de basse température, le matériau pourrait prendre beaucoup de temps à sécher, en imposant un long retard dans les délais de construction, ou pire de promouvoir la croissance de moisissures dans le matériau ou les structures de bois. Trop d'eau pulvérisée pourrait aussi peser sur le matériau, ce qui rend l'affaissement du matériau et provoquant des vides dans la cavité murale installée. Une solution à ces problèmes serait d'inclure un additif qui améliore le séchage et / ou renforce le matériau, ce qui réduit sa posologie initiale de l'eau. Idéalement cet additif doit être basé sur les ressources naturelles ou renouvelables, afin de préserver le caractère écologique du matériau. Il est avec cette stratégie à l'esprit que le projet de cette thèse a été conçu. Les objectifs de ce projet sont les suivants:

• Étudier les propriétés d'isolation en fibre de cellulose

• Étudier l'influence du dosage en eau sur les caractéristiques de performance de la matière première

• Caractériser additifs potentiels pour être utilisé avec la ouate de cellulose par voie humide, et étudier leur influence sur ces caractéristiques et d'optimiser la nouvelle formulation.

• Modéliser la performance hygrothermique dynamique de diverses formulations de la ouate de cellulose dans différentes conditions climatiques.



Figure 1: Ouate de celulose

Les travaux de ce projet de recherche est la collaboration du Laboratoire de Chimie Agro-Industrielle (INP-ENSIACET) avec la société française de conseil Greenbuilding (le coordinateur principal du projet). Il est financé en partie par des subventions de l'Association Nationale de Recherche et Technologie (ANRT), ainsi que Paris Région Entreprises. Le matériel a été fourni par SOPREMA.

Chapitre 1

Comme il a été démontré par la littérature disponible, le secteur du bâtiment a un effet important sur les émissions mondiales de CO2 et la consommation d'énergie. Deux méthodes pour réduire l'impact environnemental du secteur bâtiment sont l'augmentation de l'exigence d'isolation des bâtiments nouveaux et existants, et l'utilisation de matériaux de construction alternatifs composés de matériaux naturels ou recyclés avec une faible énergie intrinsèque et de l'impact écologique. La ouate de cellulose est un matériau d'isolation écologique innovant qui présente des caractéristiques similaires en termes de confort thermique et la performance à ses homologues non renouvelables. Néanmoins, dans son état actuel, il présent quelques inconvénients par rapport à moins de matériaux d'isolation écologiques et a montré la nécessité d'optimisation et de développement.

D'autres recherches doivent se concentrer sur l'étude et la résolution des problèmes avec les propriétés et les performances du matériau.

Bien que la recherche disponible actuellement présente en détail les propriétés de la ouate de cellulose comme installé par les méthodes sèches (qui ont toujours été les techniques d'installation les plus répandues), il y a peu de renseignements à jour sur la méthode de pulvérisation par voie humide. Un travail a montré le principal problème avec la méthode de pulvérisation par voie humide: les temps de séchage assez long en fonction du période de pose.

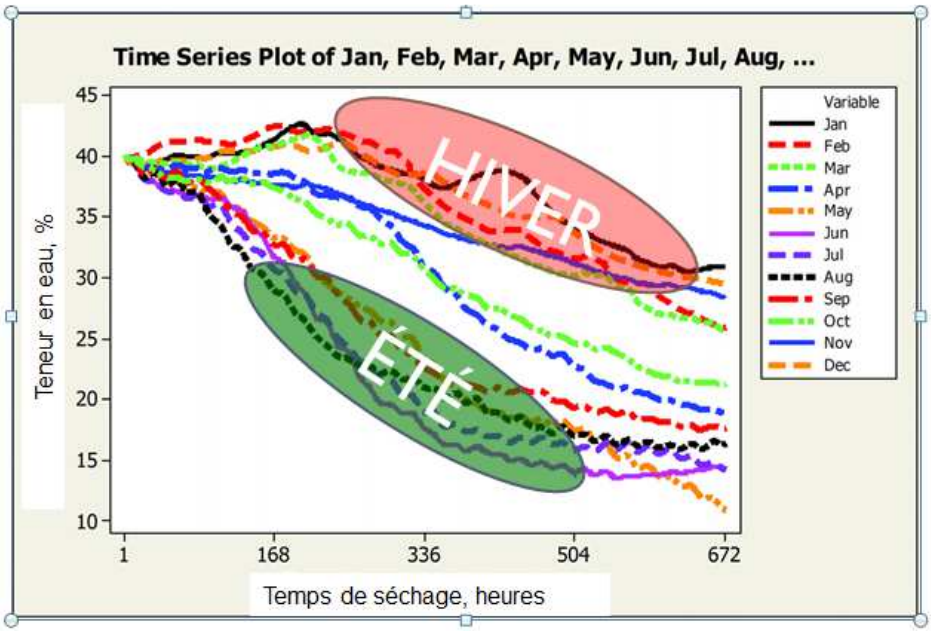


Figure 2: Séchage de la ouate de cellulose: Mikael Salonvaara, Marcin Pazera, Achilles Karagiozis, Impact of Weather on Predicting Drying Characteristics of Spray-Applied Cellulose Insulation, ASHRAE Report, 2010

L'étude bibliographique a également démontré la variabilité de la teneur en humidité est installé, ce qui conduit également à une augmentation des temps de séchage. Aucune recherche n'a pas encore déterminé l'effet de l'eau appliquée sur la conductivité thermique de l'isolation de cellulose, ou caractérisé les performances mécaniques du matériau lorsqu'il est appliqué avec l'eau. Les travaux de recherche actuelles sur les adhésifs composés de polymères naturels montre la variété des candidats potentiels qui pourraient être utilisées comme additif biosourcé avec isolation en cellulose pulvérisée humide. Bien qu'il existe de nombreux exemples d'adhésifs biosourcés utilisés dans la fabrication de composites de fibres naturelles telles des fibres de bois, la pâte à papier et le béton de chanvre, la fabrication de ces composites diffère de celle de la forme humide

de la ouate de cellulose pulvérisée. Plus particulièrement, les conditions spécifiques dans lesquelles en fibre de cellulose est fabriquée et appliquée va définir la nature de l'additif biosourcé à utiliser. La plupart des exemples cités ont des exigences de température, pression, et / ou matériel spécifiques qui ne sont pas compatibles avec le procédé de pulvérisation par voie humide. A titre d'exemple l'utilisation d'un polymère thermodurcissable serait irréalisable car il serait extrêmement difficile de chauffer un polymère pendant le processus de pulvérisation, pour ne pas mentionner les coûts associés à ce procédé. Donc l'adhésif candidat approprié devrait être une perte de solvant adhésif qui durcit avec l'élimination de l'humidité à température ambiante.

Les mécanismes qui définissent l'adhésion de processus sont complexes et variées. Compte tenu de l'adhérent (coton lâche comme des fibres de cellulose poreuses), le liant optimal va réagir et d'adhérer à des fibres de cellulose à travers principalement adsorption (liaisons hydrogène), avec les mécanismes mécaniques et de diffusion également lieu. Ces propriétés optimales sont détaillées plus en profondeur dans le chapitre 3.

En ce qui concerne la réduction des temps de séchage, quatre scénarios pourraient être envisagés par l'utilisation d'additifs biosourcés:

• La sorption de l'humidité et le durcissement ultérieur de l'adhésif ajouté accélère le débit d'eau d'évaporation de la ouate

• La résistance mécanique accrue de la ouate en raison de l'ajout d'un liant diminue la dose initiale requise de l'eau, ce qui rend le matériau atteindre un état sec plus tôt.

• L'augmentation de la teneur en matières solides du TPI avec l'additif ajouté à l'humidité diminue la proportion de matières solides, rendant ainsi le matériau initialement plus sec, puis avec un peu d'eau.

• La tension superficielle de l'eau réduite par l'utilisation d'un tensio-actif permet un écoulement plus rapide de l'eau à travers les pores du matériau, ce qui diminue les vitesses de séchage. L'absorption des tensioactifs cationiques à travers les fibres chargées négativement pourrait aussi avoir une influence.

Ces additifs ont besoin pour améliorer ces propriétés sans empêcher la performance de l'isolant en fibre de cellulose. Principalement la densité et la conductivité

thermique de la nécessité matérielle restent bas, mais aussi la résistance au feu et fongique de la matière ne doivent pas être affectées. Il est par ces innovations qui isolations à base de cellulose de fibres-peuvent devenir plus fréquents et de contribuer à d'autres projets de construction respectueux de l'environnement.

Chapitre 2

Plusieurs facteurs parmi les propriétés des fibres d'isolation de cellulose peuvent contribuer à leur comportement à l'eau. Sur les deux marques d'échantillons de ouate de c cellulose qui ont été testés, les résultats ont montré quelques dissimilitudes dans ces propriétés qui imposent des différences de performance de séchage des échantillons de cellulose pulvérisée humide. Ces propriétés peuvent comprendre sa composition chimique et les proportions de la lignine, la cellulose et les hémicelluloses, la distribution de taille des particules et la morphologie des fibres, ainsi que les additifs inclus. D'autres formes de caractérisation des fibres cellulosiques et leur relation avec l'eau comprennent la rétention d'eau, ont été étudiés la sorption dynamique de vapeur et calorimétrie différentielle à balayage pour déterminer la teneur en eau liée et non liée. Malheureusement, en raison de la variabilité des matières premières, en raison des différences de journaux recyclés, ces facteurs sont difficiles à contrôler et optimiser. Néanmoins, ils donnent des indications sur le comportement des fibres avec de l'eau et leur séchage ultérieur une fois appliqués.

		Univercell (traité)	Univercell (non traité)	Ouateco (traité)
ADF/NDF	Cellulose	34.7%	39.9%	45.3%
	Hémicelluloses	16.8%	17.3%	14.3%
	Lignine	20.4%	29.8%	25.3%
	Cendres	3.5%	5.4%	1.0%
	Autres	24.6%	7.7%	14.0%
Densité Tapée	Densité initiale (kg/m^3)	21	24.7	17.3
	Densité tapée (kg/m^3)	43	45.3	47.9
	Compressibilité (%)	49%	46%	64%
Schopper Reigler	(° SR)	42 (±3)	36 (±2)	42 (±3)

Figure 3: Propriétés phisicochimieques de la ouate de cellulose

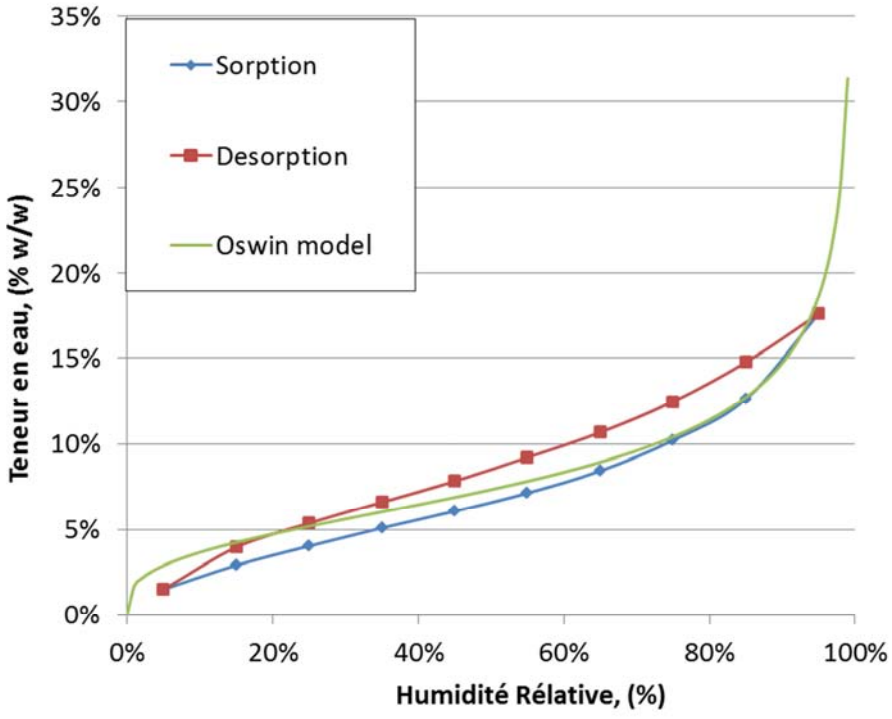


Figure 4 : Sorption d'eau de la ouate de cellulose

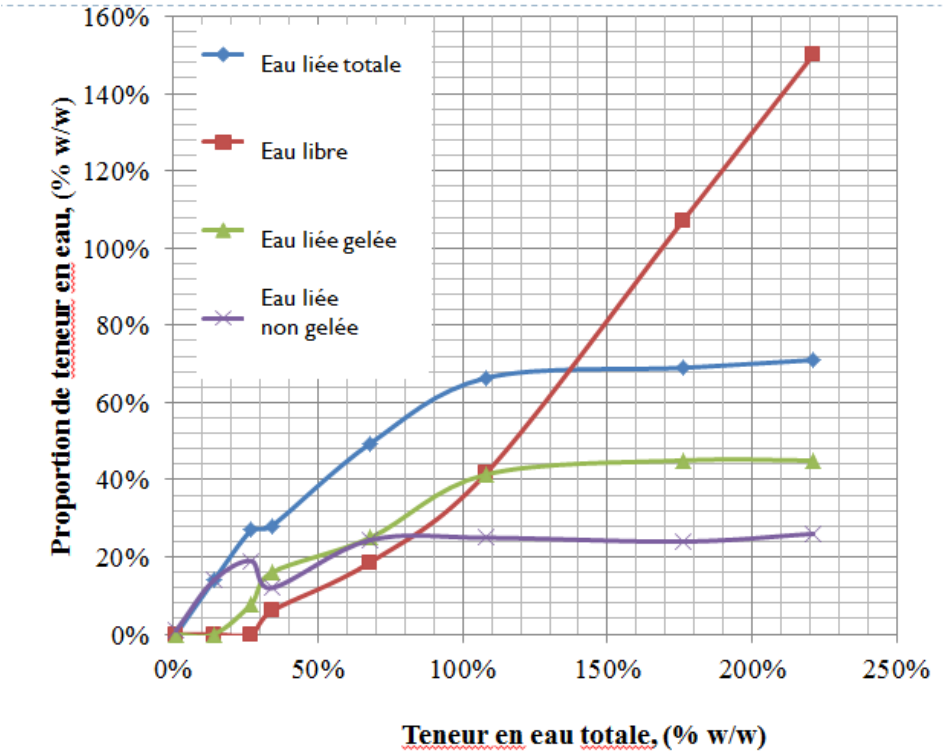


Figure 5 : Types d'eau contenue dans la ouate de cellulose

Comme les résultats ont montré, l'augmentation de l'eau non seulement retarde la construction après l'installation, mais augmente également la densité et la conductivité thermique isolant cellulosique pulvérisé humide. L'augmentation de la densité et le temps de séchage avec une augmentation de la posologie de l'eau restent des facteurs importants à considérer lors de l'application d'isolation de cellulose. Une méthode pour déterminer le comportement mécanique de la cellulose pulvérisée humide a été conçue à travers des mesures en compression. Alors qu'en réalité, les essais de compression ne représentent pas complètement le comportement des fibres de cellulose, une fois qu'elles sont projetées, elles ne donnent une indication de la façon dont la teneur en humidité installé renforce le matériau afin d'empêcher l'affaissement ou la déchirure du matériau. Une ligne de base de 14,05 kPa module d'élasticité E était définie comme propriété minimale du matériau (dans des conditions d'humidité ambiante) pour empêcher la décroche. On a constaté que l'eau appliquée non seulement densifie le matériau, mais comme les fibres gonflent et deviennent rigides au cours du séchage, l'augmentation de la résistance mécanique peut être observée. En pratique, il serait pertinent d'avoir un système de

contrôle de qualité du dosage de l'eau pulvérisée par voie humide a été mesurée dans un échantillon d'essai avant l'application sur un mur complet. Des essais mécaniques tels que celui développé dans ce travail pourrait aider à vérifier que le dosage de l'eau appliquée adaptée, le matériau peut maintenir une résistance mécanique minimale pour empêcher la décroche. Tandis que les changements dans la conductivité thermique peuvent être considérés comme négligeables, il existe toujours une perte d'efficacité thermique du matériau une fois qu'une quantité excessive d'eau a été utilisée.

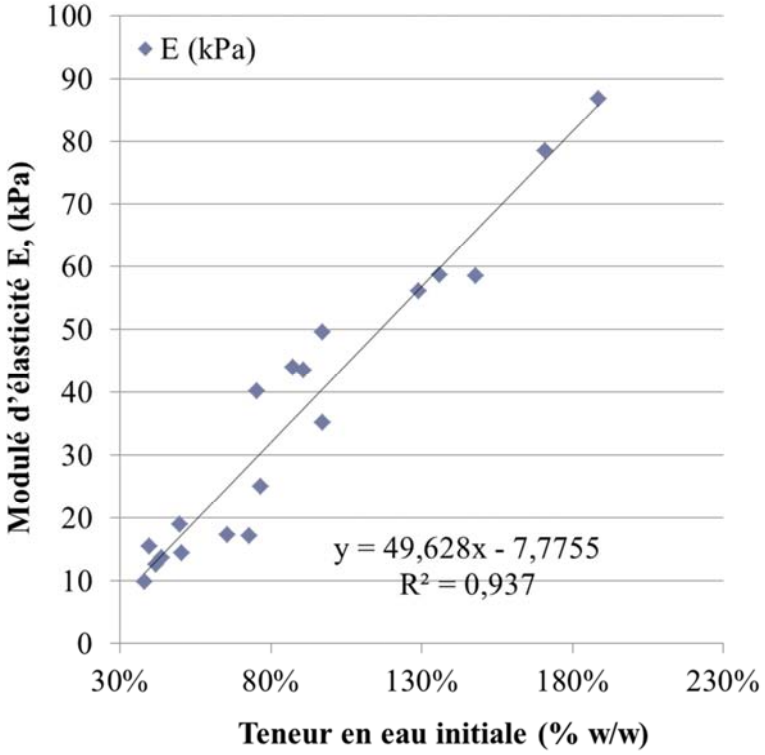


Figure 6 :Module d'élasticité vs concentration d'eau

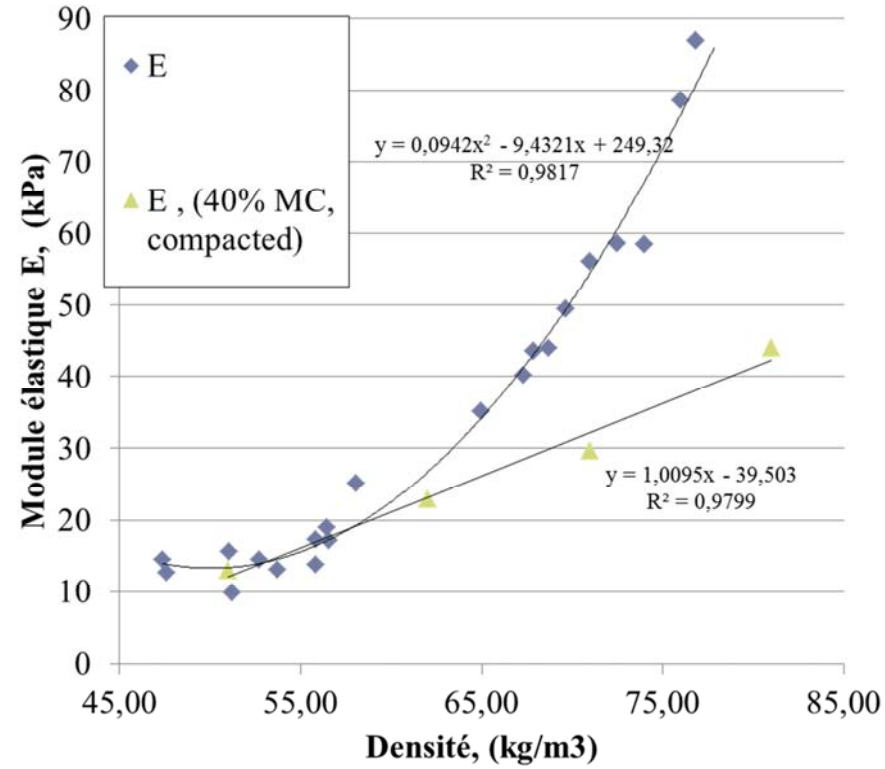


Figure 7: Module élastique vs densité

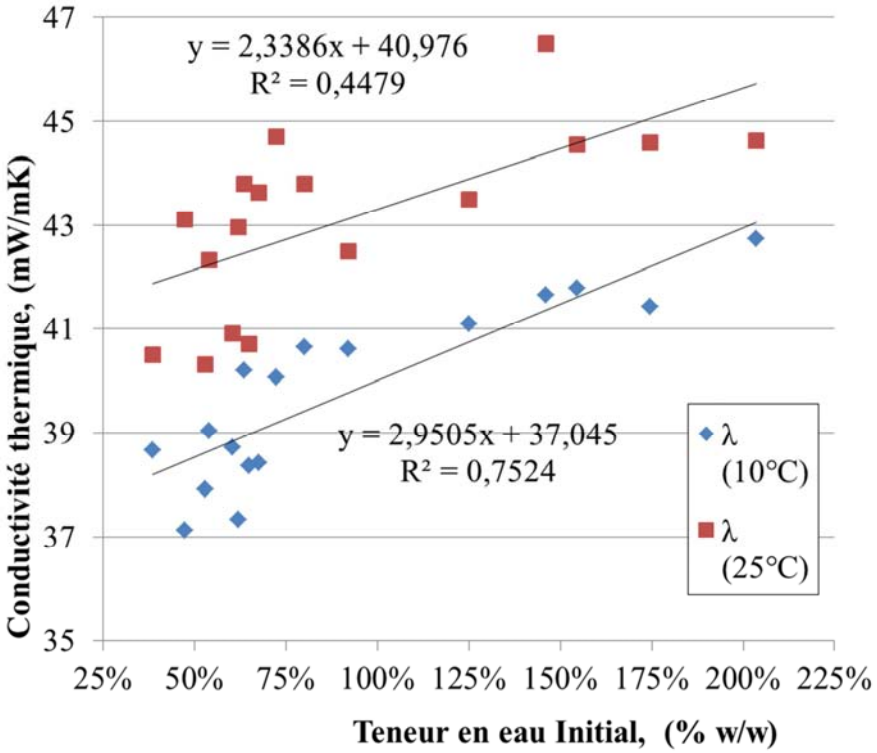


Figure 8: Conductivité thermique vs teneur en eau

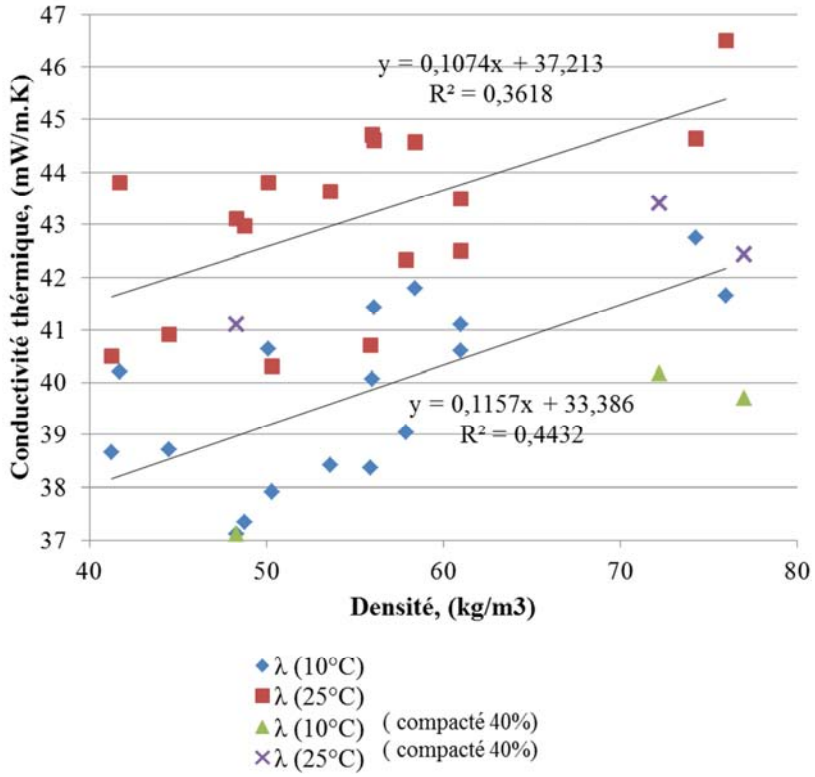


Figure 9: Conductivité thermique vs densité

Ces résultats serviront de base pour mieux comprendre le comportement de l'isolant de cellulose avec des additifs ajoutés qui améliorent son temps de séchage et les propriétés de résistance mécanique.

Chapitre 3

A partir des travaux des chapitres précédents, une stratégie a été utilisée pour trouver un additif approprié pour être incorporé à jet humide isolant en fibre de cellulose. D'abord, les additifs disponibles ont été caractérisés, en ce qui concerne leur viscosité, et leurs forces adhésives. La plupart ont montré un comportement newtonien à des concentrations faibles, avec des concentrations présentant un comportement d'amincissement de cisaillement où le seuil de viscosité pulvérisable pourrait être atteint. Malheureusement, cette promotion plusieurs liants, puisque la viscosité est une propriété inhérente des adhésifs. De nombreux additifs qui ont montré de bonnes propriétés adhésives sont trop visqueux pour être appliqué à l'équipement de pulvérisation ou, inversement, a montré une viscosité suffisante pour être pulvérisé, mais a montré une faible résistance adhésive. Une gamme de tensioactifs non ioniques, cationiques et

anioniques, des agents tensioactifs ont également été testés quant à leur tension superficielle fonction de la concentration. Les additifs conformes aux exigences d'adhésion et de viscosité ont été déterminées; après, ils ont été testés dans une plus grande échelle avec une isolation en cellulose et l'équipement de pulvérisation. D'abord, les additifs candidats ont été pulvérisés sans cellulose pour s'assurer que la quantité correcte de liquide sera appliquée.

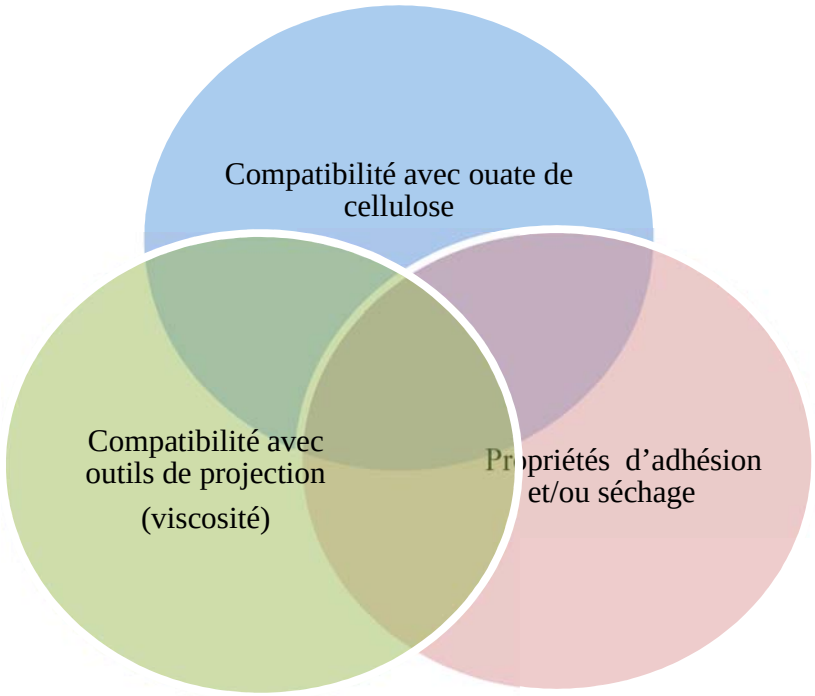


Figure 10: Propriétés d'additif récherché

Dans certains cas, l'augmentation de la viscosité des liquides de fluidification par cisaillement nécessite une pression de pompage plus élevée pour avoir le même débit. Après avoir vérifié que les additifs peuvent être traitées par pulvérisation à un débit similaire à celle de l'eau, ils ont été appliqués avec de la cellulose projetée dans les mêmes conditions que effectuées précédemment. Ils ont ensuite été caractérisés en ce qui concerne leur temps de séchage. À l'exception notable de lignosulfonates, on a constaté que pour les liants, l'augmentation de la viscosité et la formation d'une croûte de la surface ralenties évaporation de l'humidité. Le seul cas où un séchage plus rapide a été observé a été grâce à l'augmentation de la teneur initiale en matières sèches avec, en particulier, une forte concentration de lignosulfonates. Le tensioactif CTAB a été trouvé pour améliorer légèrement ces temps de séchage ainsi. Ces échantillons ont ensuite été

testés en ce qui concerne la résistance à la compression et la conductivité thermique ont été trouvés Certains échantillons pour augmenter la résistance mécanique à la teneur en humidité et densité similaire au minimum, mais la pulvérisation malheureusement n'a pas été possible en raison de la distribution hétérogène de l'humidité et un grande production de poussière. Par conséquent, après avoir examiné les additifs en ce qui concerne leur compatibilité avec l'isolant cellulosique et de son équipement, seulement lignosulfonates et CTAB ont restés comme candidats possibles. Les ectines et des tanins, qui n'a pas amélioré le temps de séchage encore montré une amélioration des performances mécaniques, pourraient être potentiellement utilisés dans la fabrication de isolants de cellulose préfabriqués.

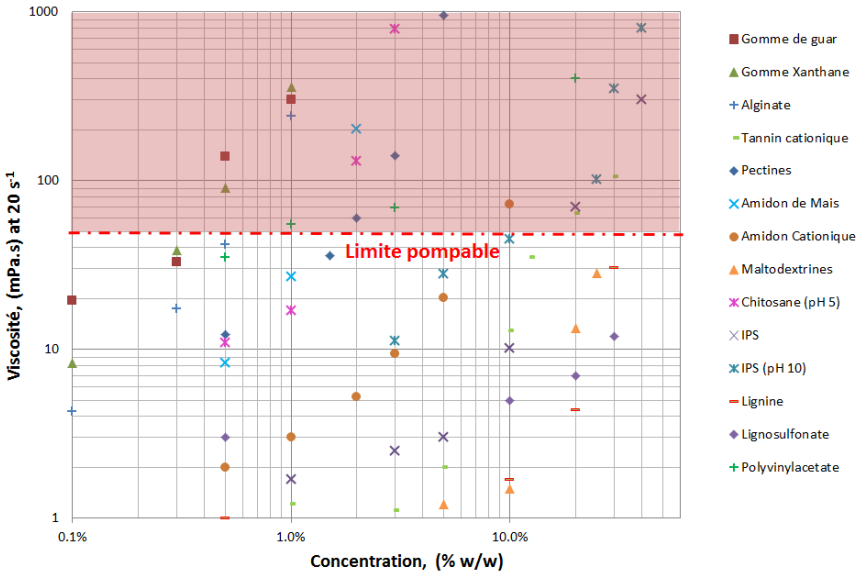


Figure 11: Concentration vs viscosité additifs biosourcés

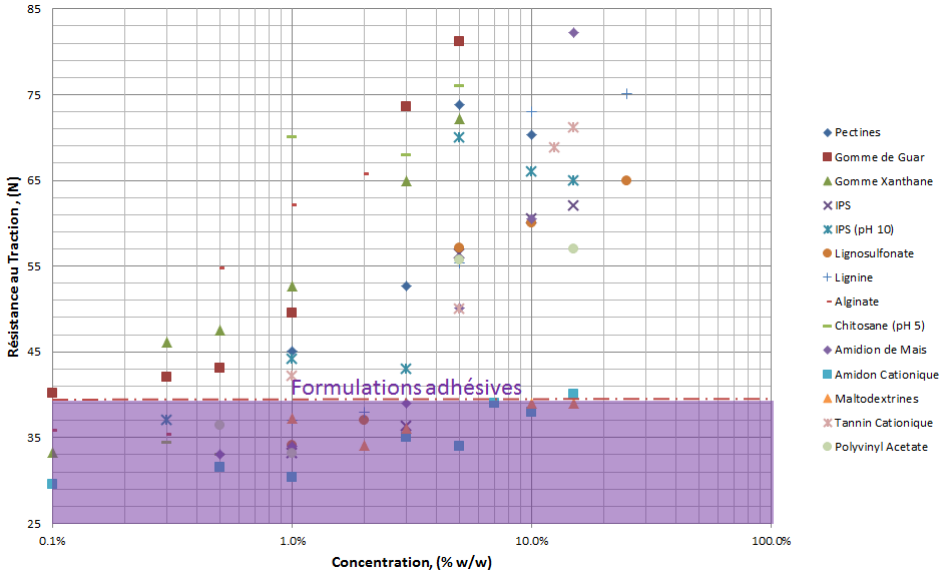


Figure 12: Concentration vs traction additifs biosourcés

Liant	Concentration Concentration (% w/w)
Pectine	1.5
Gomme de guar	0.5 (non-Newtonien)
Isolats de proteines de soja	15
Sodium Alginate	0.5
Tannin	12.5
Lignosulfonate	25

Figure 13: Additifs retenus

Enfin, les formulations ont été testées par rapport à leur conductivité thermique. Certains additifs ont été trouvés pour produire un matériau dense présentant une conductivité thermique plus faible que l'isolant cellulosique pulvérisé avec seulement de l'eau avec une densité finale similaire. Ceci suggère que l'inaccessibilité des plus petits pores par le liquide visqueux peut être bénéfique dans les matériaux propriétés isolantes.

Lignosulfonate a été trouvé pour augmenter la conductivité thermique à un niveau acceptable, seulement légèrement en diminuant sa capacité isolante. CTAB avait des valeurs de conductivité thermique très proche de celle des échantillons de référence.

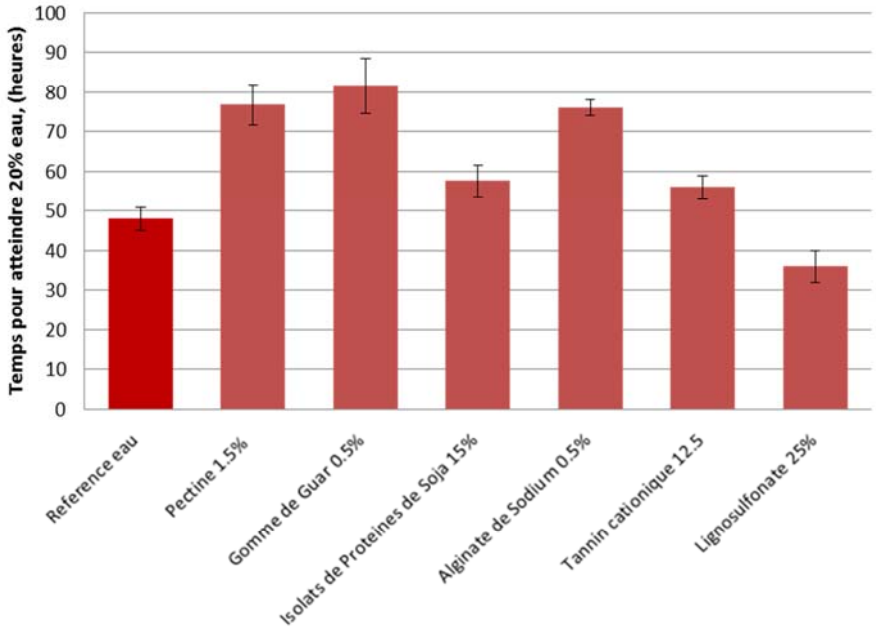


Figure 14: Séchage colles retenus

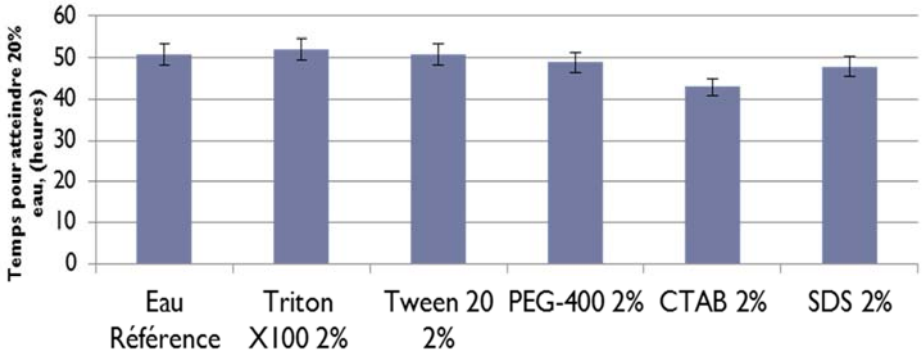


Figure 15 : Séchage Tensioactifs

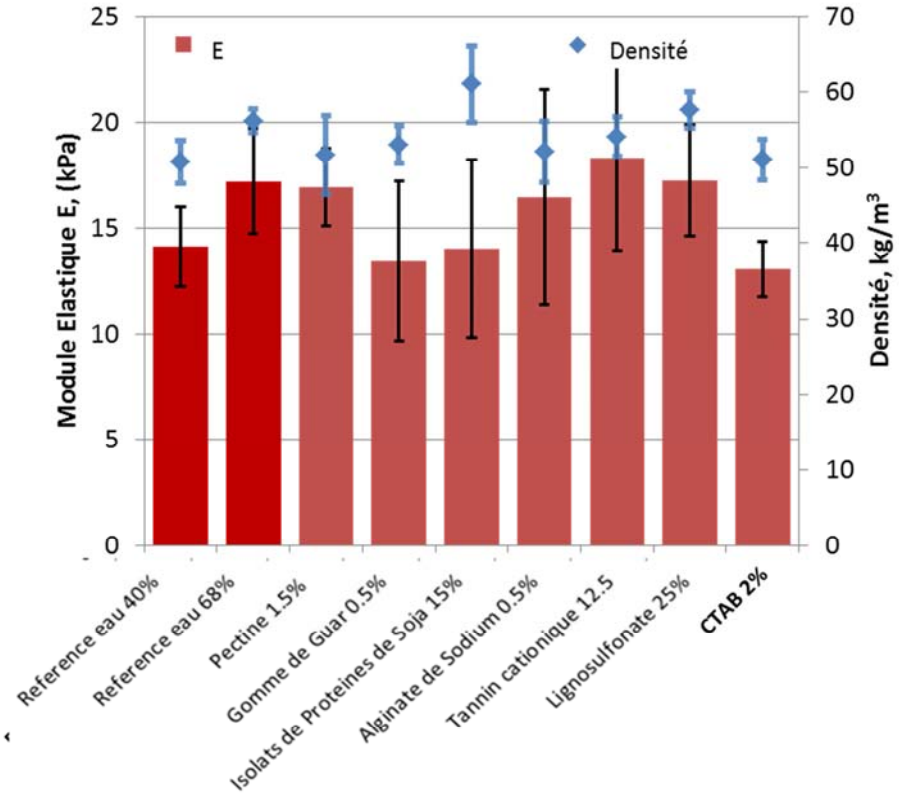


Figure 16: Mécanique formulations retenus

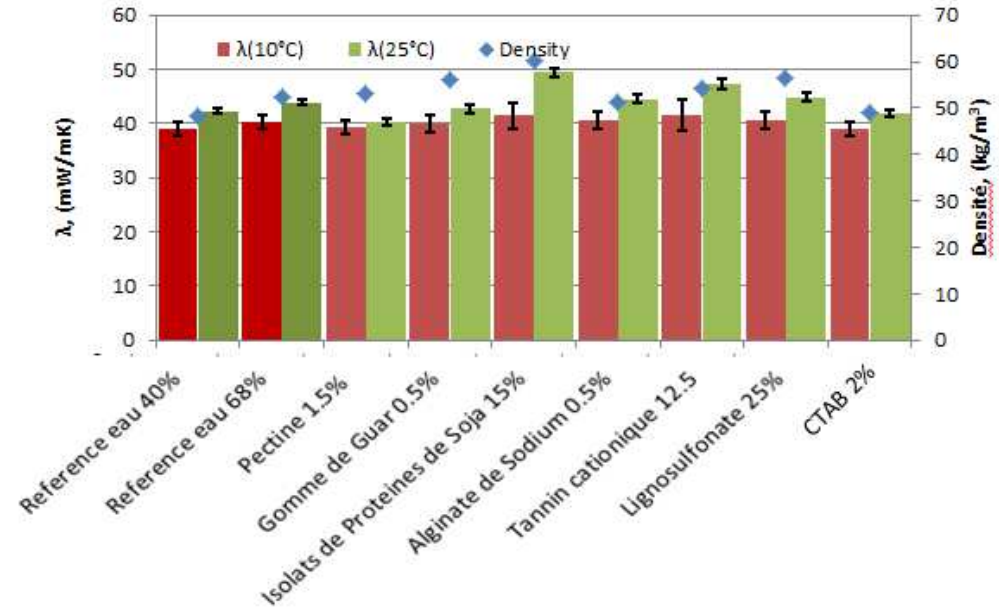


Figure 17: Conductivité thermique

Comme les essais de séchage ont été effectués uniquement en humidité relative contrôlée constante, l'influence des conditions météorologiques et épaisseur de l'isolant doit être étudié lorsque le matériau est utilisé dans des projets de construction réels. L'influence positive ou négative, le cas échéant, sur les champignons et le feu de la résistance de la matière, ainsi que d'autres facteurs pratiques, sont des questions importantes à prendre en compte aller de l'avant lors de la caractérisation du matériau isolant final.

Chapitre 4

Dans cette dernière étape du projet de recherche, les matériaux d'isolation qui incorporent la ouate de cellulose, la lignosulfonate et un tensioactif cationique (CTAB) ont été optimisés et étudiés systématiquement pour son utilisation ultérieure comme un nouveau matériau de construction. A cet égard, la concentration des deux additifs a été étudiée par rapport à leur influence du temps de séchage et les propriétés finales d'isolation. Une gamme de 19% 25% pour les concentrations lignosulfonate et et 2% CTAB respectivement se sont révélés d'être le plus bénéfiques, bien que des concentrations plus élevées puissent être utilisés si nécessaire, avec l'inconvénient d'avoir un coût plus élevé, la densité et la conductivité thermique.

Propriété	Cellulose référence	LS 19%	LS 25%	CTAB 2%
Temps de séchage a 60% RH 25°C	51	42	38	45
Masse volumique, (kg/m3)	49.8	54.3	55.6	50.6
Conductivité thermique 10°C, (mW/m.K)	39	40.9	41.1	38.7
Conductivité thermique (25°C mW/m.K)	42.3	44.5	44.8	41.9
Résistance mécanique E, (kPa)	14.1	16.5	16.9	14.3

Figure 18: Propriétés formulations retenus

Pour réaliser la prédiction des temps de séchage de l'isolant cellulosique dans des conditions dynamiques du monde réel, les modèles de transports de la matière d'humidité ont été étudiés. D'abord, un modèle empirique simplifié a été utilisé, dans lequel la teneur en humidité peut être prédite en utilisant l'épaisseur de l'isolant de cellulose, la teneur en eau vaporisées et finale, ainsi que l'influence des additifs. Le modèle empirique fourni une stratégie pour prédire le temps de séchage en utilisant des paramètres simples à l'humidité et la température constante. Après un modèle numérique complet a été utilisé par le logiciel WUFI 5 de telle sorte que l'influence combinée de l'additif avec le temps lignosulfonate installé, le dosage de liquide et l'épaisseur du matériau d'isolation peut être quantifié en ce qui concerne le temps de séchage requis. Le modèle a donné des indications sur lequel les conditions étaient les plus et les moins favorables à l'utilisation de formulations lignosulfonate. Globalement, l'utilisation d'additifs ne serait pas recommandée lors de l'installation à haute épaisseur associée à des conditions de dosage liquides élevées, mais les temps de séchage rendent ces conditions impraticables, même sans additifs. Pour été et d'automne des climats à la dose de pulvérisation minimal de 40%, lignosulfonate est recommendé à partir des épaisseurs supérieures à 160 mm.

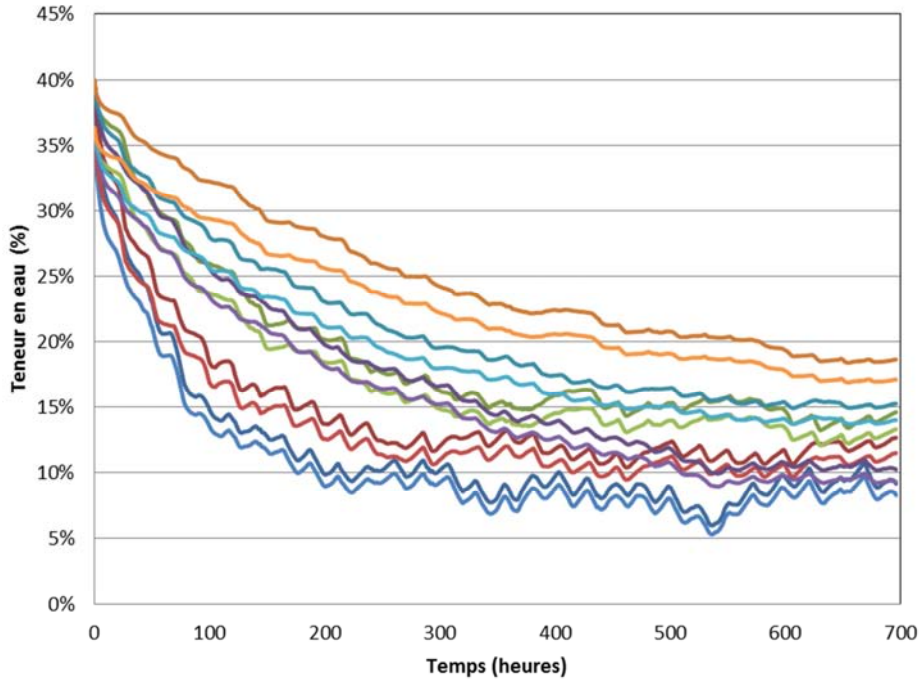


Figure 16: Impact de l’additif sur le séchage, modélisation

	Influence relative de lignosulfonate à 25% sur le temps de séchage		
	Eté	Automne	Hiver
10cm - 40% eau	23%	21%	32%
20cm - 40% eau	15%	17%	24%
10cm - 80% eau	10%	9%	5%
20cm - 80% eau	8%	7%	8%

Figure 17: Impact de l’additif sur le séchage, modélisation

Des études préliminaires en ce qui concerne le comportement au feu des nouveaux isolants à base de ouate de cellulose avec additifs ont été faites. Une première étude a été réalisée pour la croissance fongique, car additifs inclus pourraient servir de remplacement partiel pour les sels de borax inclus. Un problème avec l'utilisation des additifs de pulvérisation soit comme agents de résistance antifongique ou d'incendie, est que cela rend la charge de fournir un feu et matériau résistant à la moisissure plus dépendante de l'installateur de cellulose et non le fabricant, ce qui augmente le risque de ces complications si le matériau est mal installé, ce qui pourrait ne pas convenir à certains constructeurs. Les matériaux d'isolation candidats avec des additifs montrent une amélioration des temps de séchage et la résistance mécanique sans affecter grandement sa conductivité thermique et de la densité. Lignosulfonate était le plus efficace à cet égard, mais CTAB pourrait être utilisé comme une alternative lorsque la maitrise des propriétés de la matière est nécessaire. Une estimation initiale en ce qui concerne l'influence relative des additifs sur le prix et de l'impact écologique de la matière a été faite, ont été négligeables dans le cas de lignosulfonate, avec la variante CTAB ayant un prix similaire, mais l'impact écologique plus élevé en termes de demande d'énergie et le potentiel de réchauffement climatique. Plusieurs indications pour la caractérisation du produit final

ont été identifiées. La résistance à la corrosion, ainsi que des tests complets de résistance fongiques sont propriétés nécessaires qui doivent être définis pour la validation du matériau. D'autres propriétés comprennent: l'absorption acoustique, la résistance à l'air, la toxicité, et les tests d'odeur, entre autres. Enfin, des tests sur place pleine échelle doivent être faites sur un chantier de construction pilote pour vérifier l'efficacité des additifs dans des conditions de pleine échelle et en comparaison avec les modèles numériques.

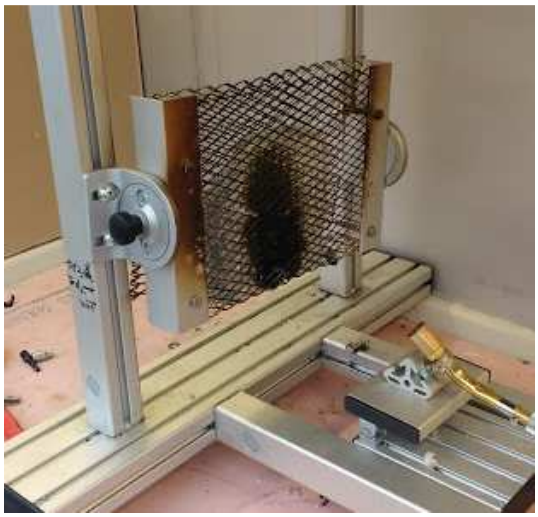


Figure 18: Mesures à la résistance au feu

Conclusions et perspectives

Pour la suite de ce travail, un test complet de séchage à grande échelle, directement dans un chantier de construction est envisagée. les compteurs d'humidité dans le matériau après sa pulvérisation, couplée à des capteurs de température et d'humidité ambiantes va permettre la surveillance en continu du séchage du nouveau matériau d'isolation et une confirmation du modèle numérique. D'autres travaux doivent également être fait sur la conformité du matériel aux normes nationales et internationales pour les matériaux d'isolation.

Au-delà de l'objectif industriel du développement du matériau isolant innovant, d'autres recherches sur l'isolation de cellulose, englobant différents domaines scientifiques, pourrait être envisagée. Alors que dans l'échelle industrielle l'optimisation des propriétés physiques et chimiques des fibres n’est pas possible, une étude en profondeur sur l'optimisation de la composition chimique et les propriétés physiques des

fibres pourrait donner des résultats qui sont applicables dans la production de papier ou de matériaux incorporant des fibres lignocellulosiques.

D'un point de vue mécanique fluide, le comportement de diffusion du liquide pulvérisé, la formation de gouttelettes et leur propagation dans les fibres de cellulose, en fonction de la viscosité est un sujet d'une étude ultérieure éventuelle. flux d'humidité en milieu poreux est un sujet dans lequel la théorie et l'expérimentation est en constante évolution, qui pourrait être appliquée à l'étude du séchage de la cellulose. Le réseau fibreux dans lequel circule l'humidité à l'intérieur de la cellulose et sa surface peut être étudié par tomographie par exemple.

Le comportement mécanique de l'isolant cellulosique pulvérisé est un autre problème qu'il faudra examiner. Etant donné que le matériau adhère à tous les bords de la cavité du mur, il est soumis aux forces de compression traction et des forces de cisaillement à partir de son propre poids. Ces forces sont de nature dynamique car lorsque le matériau sèche, le poids appliqué diminue, mais la résistance du matériau est renforcée. En outre, une fois que le matériau est sec les variations de l'humidité absorption d'humidité et de désorption à travers le fait mécanique cyclique de la performance du matériau, ce qui nécessite des tests de résistance au fluage. Ce problème est encore compliquée par le fait que l'isolant cellulosique pourrait être pulvérisé dans différentes configurations, non seulement les différentes dimensions des cavités murales qui affectent directement le poids de la matière, mais aussi comment les différents matériaux support (OSB, panneaux de fibres, panneaux de gypse, etc.) affecte son adhésion.

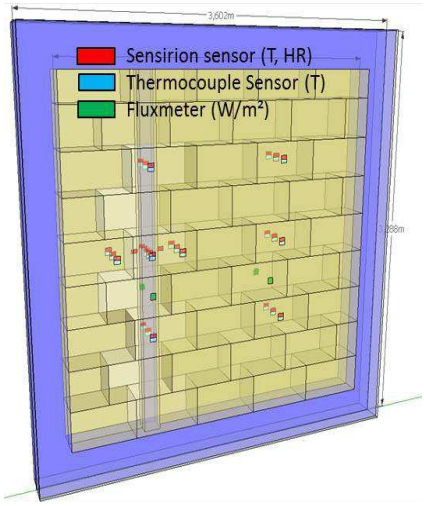


Figure 18: Dispositif expérimental envisagé

Des matériaux tels que la ouate de cellulose sont des éléments essentiels pour la réduction de l'impact environnemental du secteur bâtiment, mais leur intégration dans des projets de construction est toujours moins favorable que les matériaux traditionnels. Il est seulement par itération, l'innovation et la recherche que les matériaux de construction qui incorporent des fibres naturelles et recyclées deviendront plus fréquente, le standard et non l'exception.

PUBLICATIONS :

Hurtado, P. L., Rouilly, A., Vandenbossche, V., & Raynaud, C. (2016). A review on the properties of cellulose fibre insulation. *Building and Environment*, *96*, 170-177.

Hurtado, Pablo Lopez, Antoine Rouilly, Christine Raynaud, and Virginie Vandenbossche. "The properties of cellulose insulation applied via the wet spray process." *Building and Environment* 107 (2016): 43-51.