

Dynamic behaviour of bio-based and recycled materials for indoor environmental comfort

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Abstract

UK construction industry contributes 120 Mt of waste every year. Bio-based building materials may be a solution for this problem, as they combine re-use and recycling abilities together with hygroscopic characteristics, leading to buildings energy savings. For the first time, the dynamic response to hygrothermal changes of bio-based materials is examined in terms of Moisture Buffering Value (MBV), dry/ wet thermal conductivity, microstructure, density and latent heat through daily cycles. It is shown that MBV is a useful tool for characterisation but needs to be combined with the shape of the change in mass of the final hygrothermal cycle. Mastering this is required to obtain significant improved indoor environment quality in buildings. Ten samples of bio-based insulation materials and one thermoplastic recycled polymer were analysed (wool, hemp, saw mill residue, wood, straw, cork and polyethylene terephthalate). Saw and wool are the most promising, as materials exhibit dynamic response to hygrothermal changes. Only half the amount of samples revealed equivalent efficient moisture transfer to be able to desorb the adsorbed quantity of water. Latent heat of vaporisation and condensation tests led to the conclusion that samples of wool and saw mill residue can qualify as bio-based materials for 'green' panels.

Keywords: bio-based materials, insulation, hygrothermal, latent heat, moisture buffering value (MBV)

1. Introduction

As a direct response to the Kyoto Protocol coming into force in 1997, (and the first of its kind as a legally binding commitment to combat climate change between 2008-2012) the Climate Change Act 2008 (H.M.Government, 2008) set a benchmark for the United Kingdom's (U.K.) government to reduce carbon emissions. This Act outlines an 80% reduction of emissions from baseline levels set in 1990 and provides a set of 5-year budgets till 2022 (with a guaranteed reduction in emissions by at least 26% before 2020).

Despite this, between 2014 and 2015 the U.K.'s carbon footprint actually rose by approximately 2% (H.M.Government, 2018) - highlighting the urgent need for more environmentally considerate construction techniques and materials. As highlighted by (Giesekam, Tingley and Cotton, 2018), within 2014 the construction industry and built environment sector as a whole emitted around 183.5 MtCO₂e (Giesekam and Pomponi, 2017)

– where approximately 25% was linked to embodied emissions . There is a clear conundrum facing the U.K. construction industry in terms of balancing an obligation to climate change in addition to meeting the requirements and demands of the building industry (Atkins, ICE and ITRC, 2016; Krausmann et al, 2017). As emissions continue to rise, the U.K. government has shown commitment to combat them via various mechanisms such as Construction 2025 (H.M. Government, 2013a), Construction Strategy 2016-2020 (H.M. Government, 2016) and industrial initiatives via the launch of Environmental Product Declarations (Passer et al, 2015) and Infrastructure Carbon Review (H.M. Government, 2013b). Further European commitment to reduce carbon emissions and utilization of bio-waste has been posed by the European Commission’s bio-economy strategy of 2012 (European Commission, 2017) where all member states of the European Union (EU) aim to reduce their emissions by 90% by 2050 (European Commission, 2011).

This mandate to reduce carbon emissions has given rise to the use of bio-based materials as structural and non-structural components in construction. However, the hygroscopic characteristics of these materials are not yet fully understood, limiting their optimisation and further adoption by the construction industry. Bio-based materials are superior to conventional construction materials (such as concrete or brick) due to their relative simplicity, abundance and ability to mimic and if not better the equivalent fossil fuel based materials (Jones and Brischke, 2017) (see Figure 1). Despite their low embodied energy, bio-based materials themselves have drawbacks due to the lack of European legislation surrounding embodied energy. Without sufficient regulation, the actual benefits of using such materials are much less measurable and classifiable (Scarlat et al, 2015).

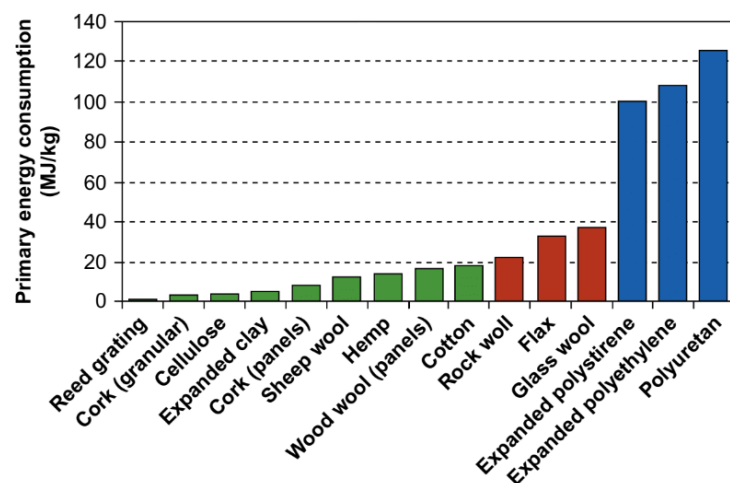


Figure 1. Primary energy consumption variation of bio-based and fossil fuel based materials (Jones and Brischke, 2017).

Despite the legal pressures to reduce emissions within the U.K., Part L of the *Building Recommendations* are still yet to highlight and focus on the legislative significance of embodied energy and have tightened requirements for operational energy (Tingley and Davison, 2011).

U.K.U.K. Whilst there is limited legislation to demonstrate the benefit of reducing embodied energy, more sustainable building materials have been developed in order to align with environmental pressures. ‘Green panels’ are a concept that has been developed as an

ecological solution as a passive method of controlling the interior hygrothermal environment and thermal within buildings. The desire for a new, environmentally friendly insulation product is more prevalent than ever before. Research into the creation of green panels is relatively limited but extremely varied due to wide and varied range of bio-fibres that are available and have the ability to grow in different climates and habitats all around the world.

When exposed to interior surfaces, bio-based products, (most noticeably insulation materials) contribute positively towards improving indoor air quality (IAQ). Within an interior surface, different house hold nuisances thrive at different indoor Relative Humidity (RH) levels, which contribute to the deterioration in IAQ – this is demonstrated in Figure 2. The hygroscopic nature of bio-based products allows for the adsorption/ desorption of water vapour into their porous structure, in dynamic equilibrium with their surrounding environment, by doing so creating a hygric buffer. This enabled the fluctuations in the hygrothermal environment to be kept to a minimum thus, aiding towards improving IAQ and also reducing the energy requirements of air conditioning (Palumbo et al., 2018) (Lawrence et al. 2013). In addition to bio-based products, other naturally occurring materials such as earth can also be attributed to these unique hygrothermal management characteristics. The quantity of moisture accumulated is material specific and depends on the RH and the temperature of the environment (Palumbo et al., 2016) and by controlling these values it is possible to accurately track the adsorption/ desorption characteristics of bio-based materials.

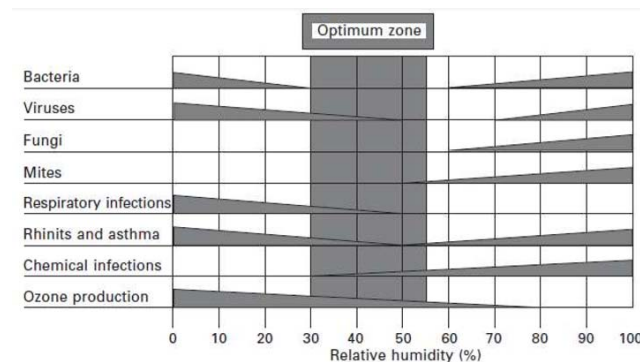


Figure 2. The optimum zone for RH against different types of house hold nuisances (Simonson, Salonvaara and Ojanen, 2001).

Generally, 'green' insulation panels can be realized by employing 'Phase Changing Materials' (PCMs) as outlined in (Cui et al, 2015) and (Baetens, Jelle and Gustavsen, 2010) and bio-based composite materials as outlined in (Laborel-Préneron et al, 2016). Due to their large specific heat capacity, PCMs are able to increase thermal comfort as they have the ability to regulate indoor temperature (Al-Saadi and Zhai, 2013). Yet the energy required for the material to change phases i.e. superheating limits their application (Chen and Qin, 2016), enhancing the need to optimise bio-based composites. Bio-fibre composites can be developed using agro-waste which is widely available. Internal wall bio-based panels optimisation has been studied by other researchers (Arnaud, 2009; Pavlik et al, 2011; Latif et al, 2016; Latif et al, 2018). However, the knowledge on the passive mechanisms and the hygroscopic characteristics associated to these materials is not fully mastered.

The inability for a material to ineffectively adsorb and desorb at the same rate and that the material is beginning to reach its saturation point, this phenomena is known as hysteresis (Lelievre, Colinart and Glouannec, 2014). In addition to this, the samples that are strongly

hygroscopic will be affected the most which would demonstrate the effect of hysteresis more prevalently (Feng et al, 2015). These results also agree with work by (Hall and Allinson, 2009) where there is a clear relationship linking saturation levels and an increase in thermal conductivity.

This paper demonstrates preliminary research which aims to use bio-based materials and recycled materials currently available on the market within the U.K., and characterise their behaviour at high levels of RH (non-optimum zone from a hygrothermal perspective). The studied materials will be case-tailored to create a 'green panel' which is envisaged to regulate a localised hygrothermal environment for buildings refurbishment. By intrinsically quantifying and classifying the optimally performant types of bio-based materials, an optimised basis for the creation of the 'green' panel will be provided.

2. Experimental Procedure

2.1. Materials Characterisation

Ten samples of natural bio-based insulation materials and 1 thermoplastic polymer were analysed: 4 different types of Wool insulation, Hemp, Wood Wool Board (WWB), Saw Mill Residue (SMR), Wood Fibre (WF), Straw, Insulated Cork Board (ICB) and Polyethylene terephthalate (PET). All materials are currently available on the market within the U.K. and the properties of each type of bio-based material are listed in Table 1.

Table 1. Materials characteristics.

| Sample Name | Sample Thickness (mm) | Density (kg/m ³) | Thermal Conductivity (W/m.K) |
|-------------|-----------------------|------------------------------|------------------------------|
| Wool 1 | 65 | 18 | 0.039 |
| Wool 2 | 75 | 31 | 0.035 |
| Wool 3 | 50 | 45 | 0.04 |
| Wool 4 | 40 | 30 | 0.039 |
| Hemp | 50 | 25 | 0.04 |
| WWB | 15 | 8 | 0.065 |
| SMR | 55 | 50 | 0.038 |
| WF | 60 | 145 | 0.041 |
| Straw | 60 | 200 | 0.0397 |
| ICB | 65 | 120 | 0.04 |
| PET | 10 | 13 | 0.04 |

2.2 Testing Procedures

2.2.1 Surface Morphology

Once samples were selected, the surface morphology and material characteristics were interrogated at micro-level. Examination was conducted by using a Scanning Electron Microscope (SEM) Inspect S.

2.2.2. Density

To be able to compare the effects moisture has on the samples both the dry and saturated density of the materials were calculated as per EN1015-6(CEN, 1999). For each type of material, 3 specimens were analysed. Density of saturated materials was determined by immersing samples in water and periodically recording weight gain, the samples were immersed until full saturation.

2.2.3. Moisture Buffering Value (MBV)

The Moisture Buffering Value (MBV) is a manifestation of the ability of a material to efficiently adsorb and desorb moisture in a dynamic hygrothermal environment. Samples were exposed to cyclic step changes of RH between 75% and 53% every 8 hours and 16 hours, respectively, at a constant temperature of 23°C in accordance with both NORDTEST protocol (Rode et al, 2005) and ISO 214353 (ISO, 2008) – Figure 3 demonstrates the step change in RH over a single 24 h cycle.

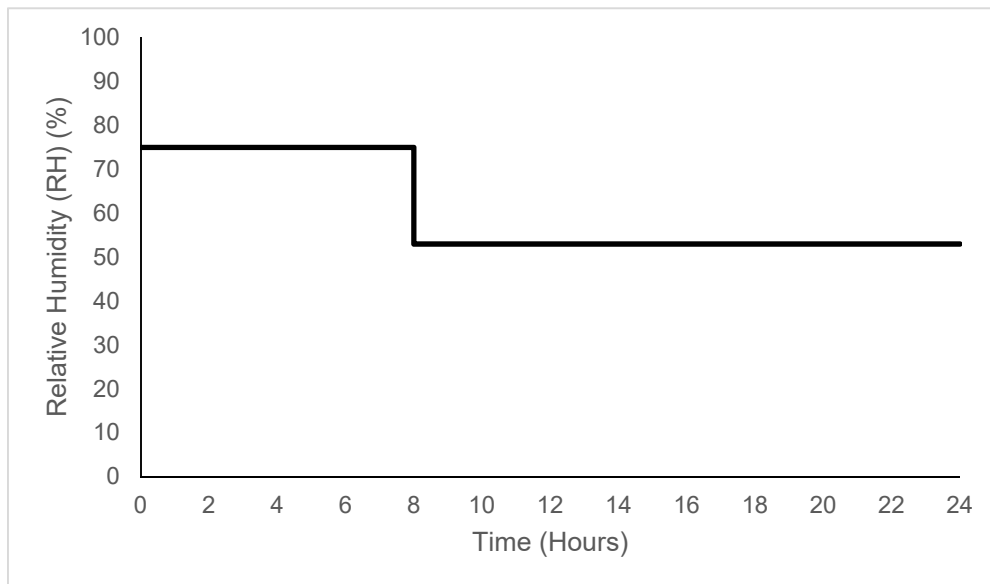


Figure 3. Step change in RH within climatic chamber over a singular, 24 h cycle

In order to determine the Moisture Buffering Value (MBV) the following equation was adopted (Equation 1):

$$MBV = \frac{m_a - m_d}{A \Delta \phi} \quad (1)$$

Where:

m_a = Mass of sample at end of moisture adsorption stage (g)

m_d = Mass of sample at end of moisture desorption stage (g)

A = Exposed surface area of sample (m^2)

$\Delta \phi$ = Difference in RH between adsorption and desorption stage (%)

Apart from satisfying the two test conditions, the adopted hygrothermal conditions mimic a residential household and the occupant's behaviour within the U.K. Samples were placed horizontally within the climatic chamber and wrapped in aluminium foil tape. All samples had a singular exposed surface area 0.01m^2 (where other surfaces were covered in aluminium foil tape).

2.2.4. Thermal Conductivity

Thermal conductivity values of the as-received and moisture-saturated samples was recorded. This was carried out by employing a thermal conductivity meter (ISOMET) that features a 60mm diameter circular contact probe (see Figure 4). Thermal conductivity measurements were conducted at laboratory conditions at 21°C and 54% RH.

Thermal conductivity was measured with the materials in three differing states: dry, saturated and how the samples dynamically react to a changing hygrothermal environment within a climatic chamber. The thermal conductivity of the samples was recorded during at distinct time intervals of 0, 8 and 24 h during the absorption and desorption phases.



Figure 4. An image of saturated bio fibre thermal conductivity reading.

2.2.5. Temperature evolution during transient behaviour

In order to detect the temperature changes within the samples, thermocouples were located on both the surface and at 50% of the depth of the sample (see Figure 5a and Figure 5b). The employed thermocouples and data loggers are functional at temperature ranges of -40°C to 260°C and -250°C to 1370°C respectively, with a maintained resolution of $\pm 0.04^\circ\text{C}$.



Figure 5a. Bio-based samples with thermocouples attached.

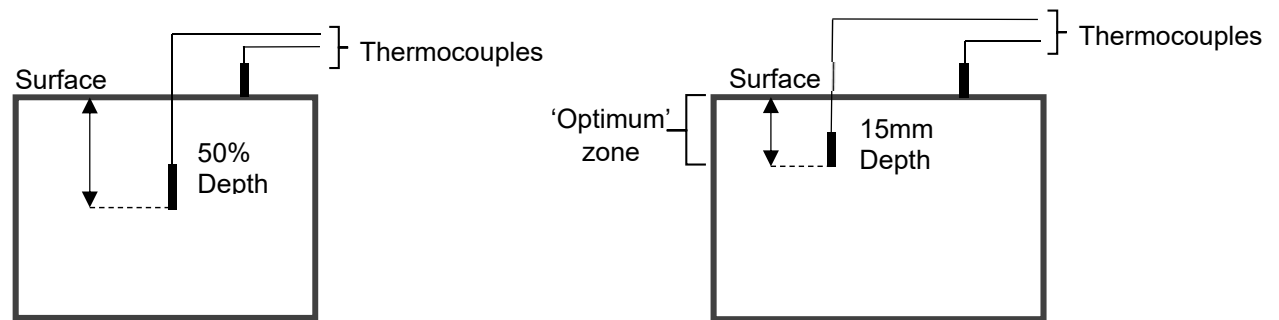


Figure 5b. (L) 'Run 1' thermocouples arrangement. Figure 5c. (R) 'Run 2' thermocouple arrangement.

Samples within 'Run 1', were exposed to 10 cycles of 24 hours, where temperature values were recorded every 30 sec time intervals. The selection of 10 cycles was chosen as it was the assumed minimum number for a representative analysis of hygrothermal behaviour of each sample, this difference is limited to below 15% between MBV values. According to (Holcroft, 2016) and (Padfield, 1999), there appears to be an optimum zone within the sample for detecting temperature changes. This area is an approximately 15 to 16mm thick material zone that start from the surface of the sample towards its core. (Padfield, 1999). This 'optimum zone' is hypothesised to be the active layer of the sample (McGregor et al, 2017). For the next cycle of testing ('Run 2'), samples were exposed to 22 cycles and scanned with thermocouples on both the surface and at a depth of 15mm (see Figure 5c), where 22 cycles represents the final stabilisation of samples.

3. Results and Discussion

3.1. Density of the materials

The density values of the samples are illustrated in Figure 6a and 6b. When comparing density data from both dry and saturated samples, it is evident that moisture increases significantly the density of the sample. This is attributed to voids or air gaps between fibres within the bio-based material are filled by water molecules rather than free flowing air. Low density values of dry samples indicate that the material is permeable enough as bonding sites for water molecules are readily available.

As can be seen in Figures 6a and 6b, it is evident that the density values of Wool 1,2,3 and 4 exhibit a substantial increase when samples are saturated. It has been hypothesised by Swift and Smith (2001) that this could be attributed to the microscale structure of wool itself and the hygrothermal conditions the sample is exposed to. Swift and Smith (2001) highlighted that depending on the hygrothermal conditions, the lipid layers that are disordered and dynamic may change positions. Therefore as a direct response of being saturated, lipid layers may change positions and when in contact with water may swell to as many as 7 times of their original size. ICB is the material that exhibited the lowest dependency on changes between dry and saturated states. This could be attributed to the extremely slow water adsorption of

water into cork cells in comparison to other bio-based materials for example: wood fibres(Rosa and Fortes, 1993).

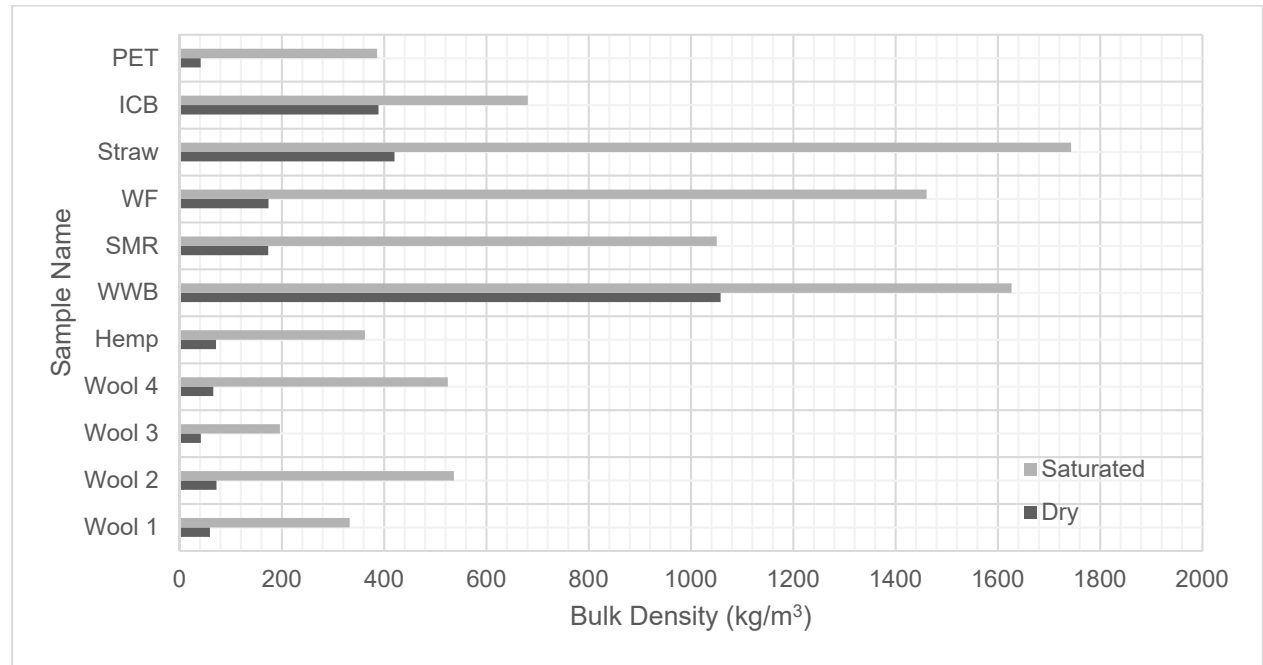


Figure 6a. Bulk density variation of samples in dry and saturated states.

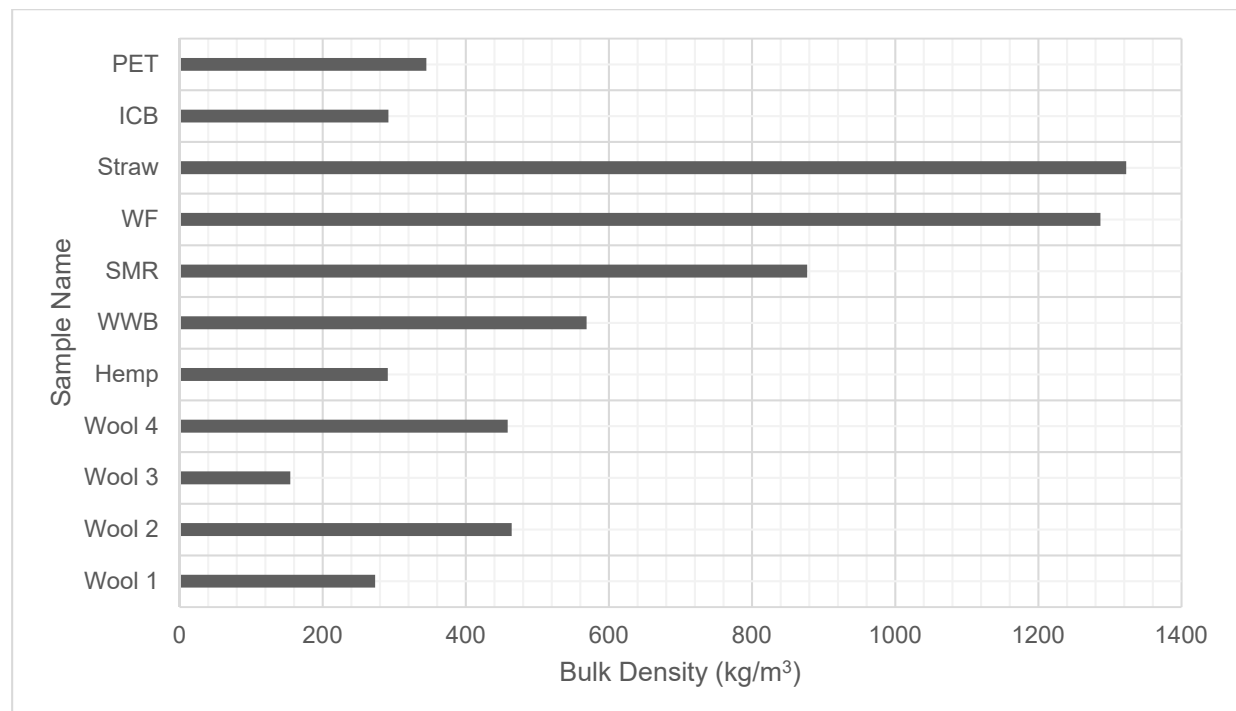


Figure 6b. Difference between saturated and dry density of examined materials.

3.2. Moisture Buffering Value (MBV)

The MBV of the examined samples were determined for each individual cycle. The MBV for 10 cycles is illustrated in Figure 7. MBV measurements were taken manually and due to accessibility issues to the climatic chamber over weekends, cycles 4, 5 and 6 have been excluded from Figure 7.

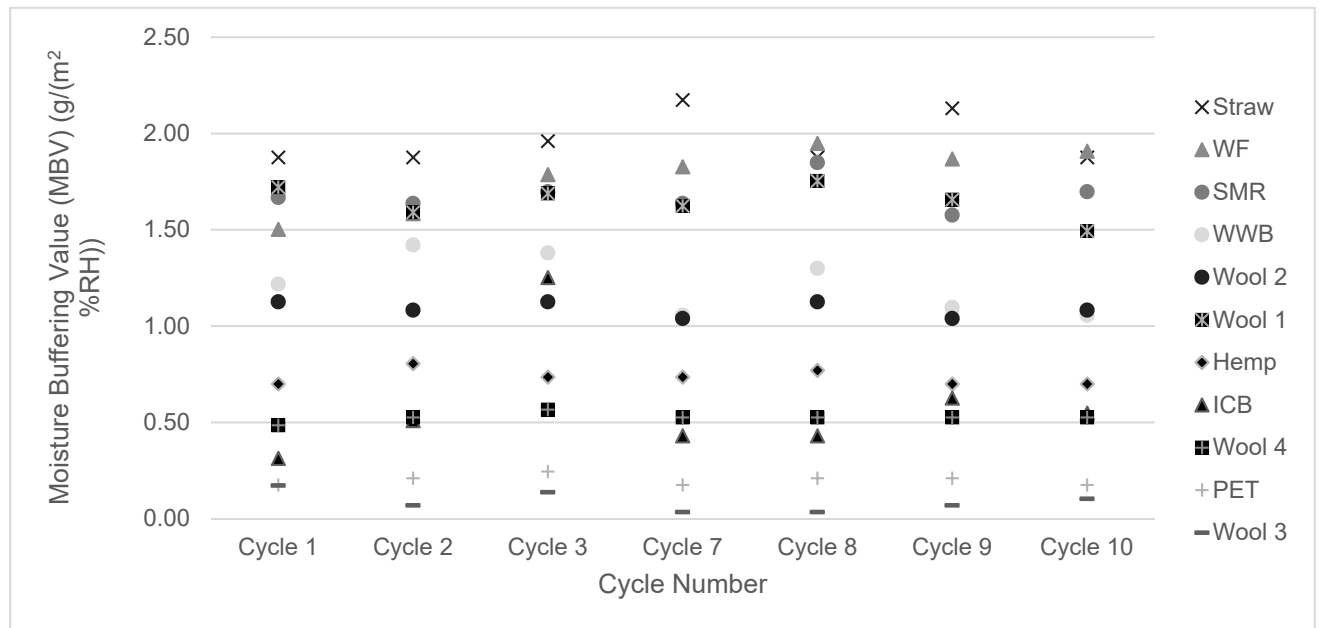


Figure 7. MBV variation of each sample per cycle

In Nordtest Protocol (Rode, 2005) test, materials must be in a quasi-steady state, demonstrated by a variation in mass of less than 5% for 3 consecutive cycles. Due to the inherently variability of bio-based and recycled materials, all samples except for ICB, Straw, Wool 1, Wool 4, SMR and PET are in a quasi-steady state within 10 cycles. This is demonstrated within Figure 7 as it demonstrates MBV values fluctuation over the course of the hygrothermal cycles. It may be postulated, that the variation between each bio-based sample is a result of a probable micro-capillary network formations created during water molecules adsorption and desorption. As this process continues, for samples that are cellulose based, transient micro-capillary networks are formed within cellulose and lignocellulose fibres. Whilst in a constant cycle of adsorption and desorption within the chamber, capillary condensation becomes more apparent (Hill, Norton and Newman, 2009). Due to their fundamental and inherently variable characteristics, different bio-based materials stabilise at different rates (McGregor et al, 2016). This explains the increased scattering within the MBV values for the different studied samples to sample as they react to the hygrothermal environment.

In addition to MBV values, the shape of the adsorption/desorption curve has received little attention. The adsorption/desorption curves can be seen in Figures 8a and 8b.

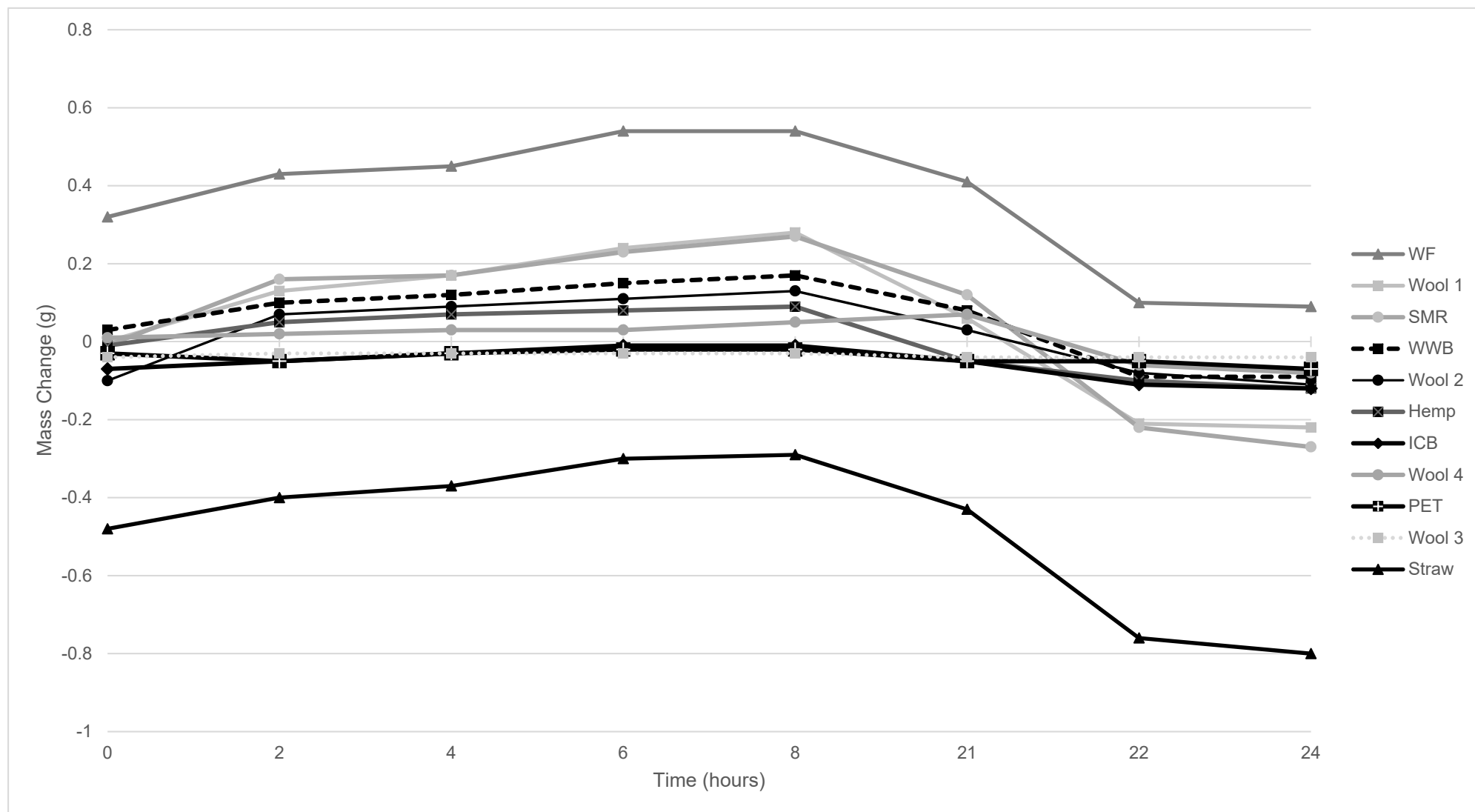


Figure 8a. Change in mass during the 7th cycle of the 10 bio-based materials and 1 recycled plastic during a 24-hour cycle.

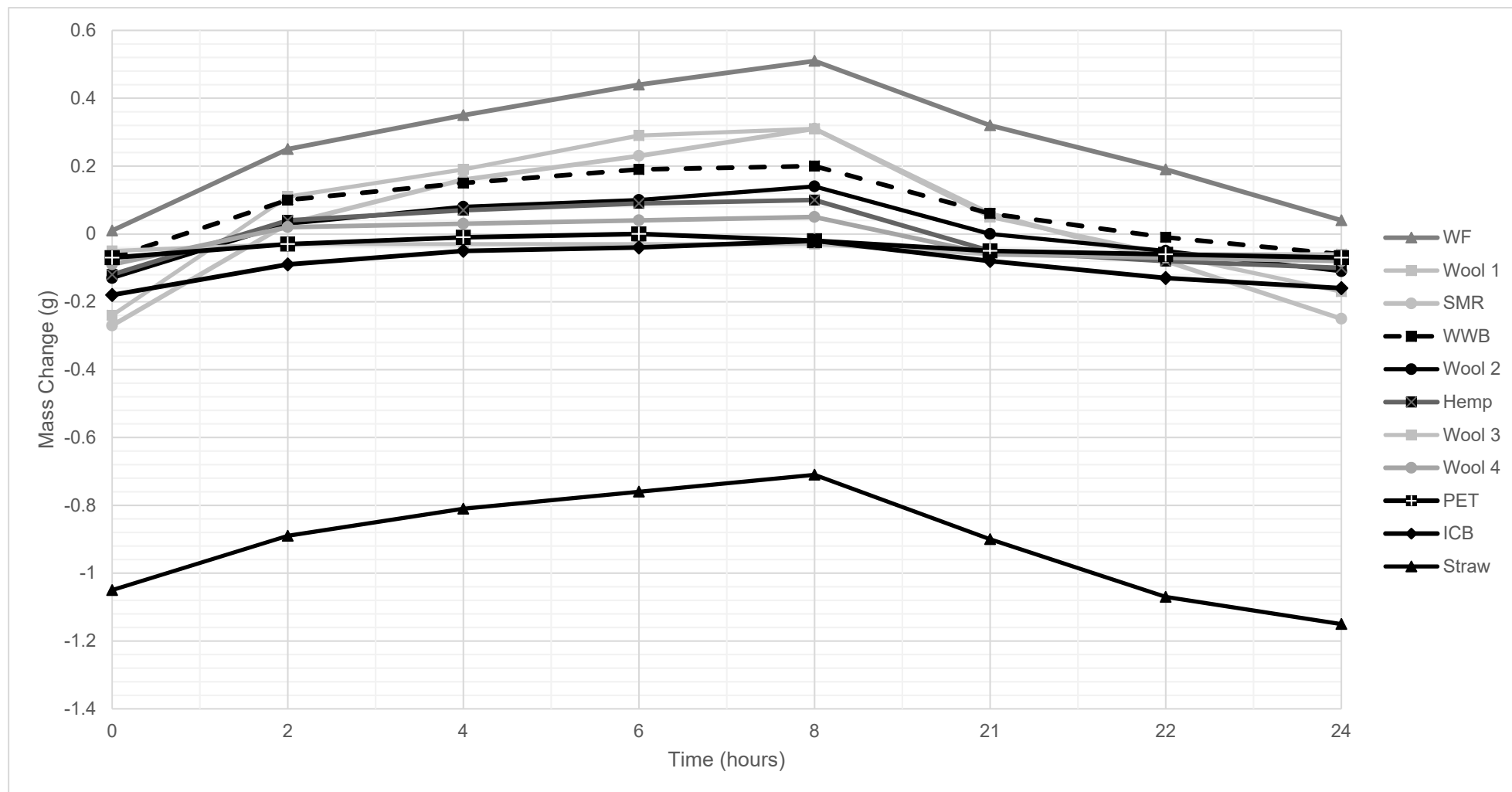


Figure 8b. Change in mass during a 10th cycle of the 10 bio-based materials and 1 recycled plastic during a 24-hour cycle.

From Figures 8a and 8b it is demonstrated that the moisture adsorption and desorption process vary for the various studied materials. The comparison of cycle 7 and cycle 10 helps to demonstrate the differentiation of shapes materials experience before they are in a quasi-steady state. Figures 8a and 8b also demonstrate the natural variation and evolution between the different materials throughout the entirety of the experiment. SMRThe figures demonstrate different adsorption/desorption curves for different materials. These curves have been categorised and the following groups have been created:

'Group 1'

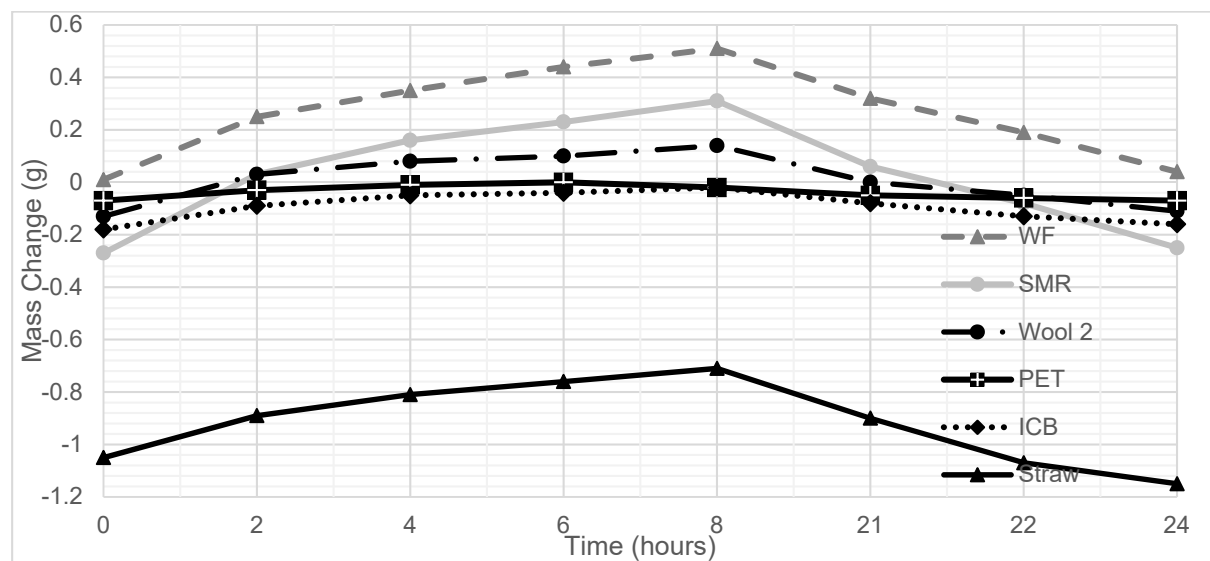
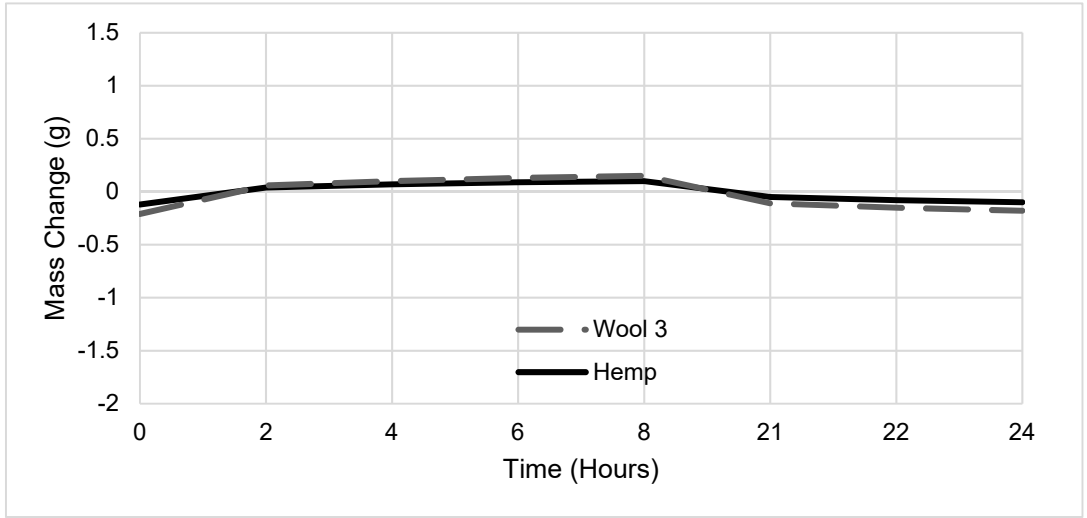


Figure 9a. Adsorption and desorption curves to show the categorisation of 6 samples within 'Group 1' (Romano et al, 2018).

Within this group, all samples initially adsorb moisture at a high rate for a period of 8 h and mass is constantly increasing (see Figure 9a). After 8 h, samples desorb moisture in a similar manner. This is mirrored in the desorption stage. Samples within Group 1, exhibit clear adsorption/desorption phases, having a peak after 8 h of exposure, which denotes the end of the adsorption phase. This adsorption/desorption curve shape corresponds to an instantaneous reaction to differential hygrothermal conditions. This behaviour clearly demonstrates the hygroscopic nature of the material having the ability to manage dynamic moisture changes. This is evidence that the material can dynamically react to differing hygrothermal environments.

'Group 2'

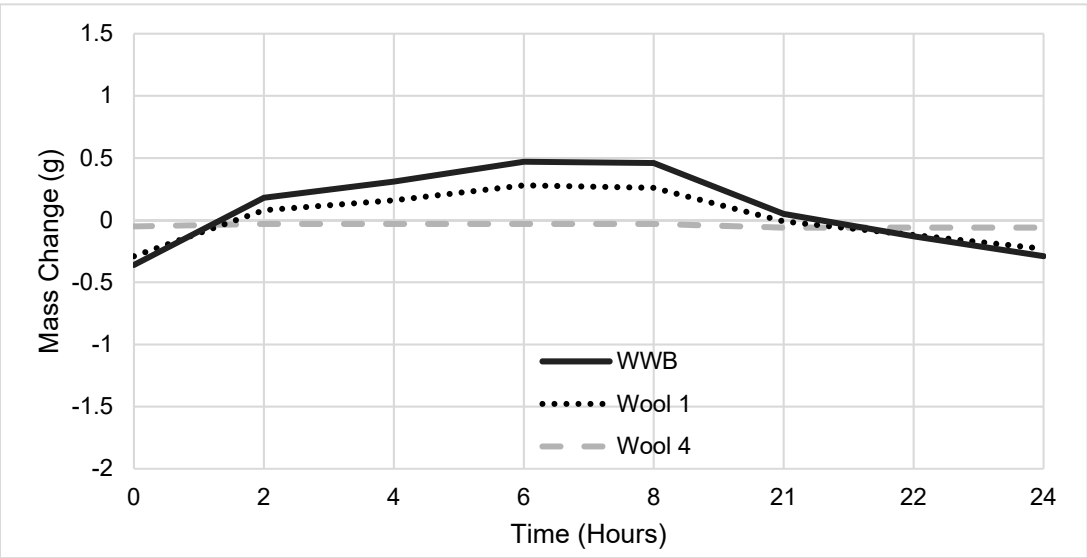
28
29



30 **Figure 9b.** Adsorption and desorption curves to show the categorisation of 2 samples within
31 'Group 2'.

32 The shape of the adsorption/ desorption curves of 'Group 2' samples exhibit an initially high
33 absorption rate (see Figure 9b) for the first 2 h of exposure. After that, a clear change in the
34 adsorption curve rate is depicted. The materials desorb moisture in a similar manner. The
35 adsorption and desorption phases in this class of materials are distinct and separated by a
36 peak in after 8 h of exposure. Although, the rate reduction is an evidence of the material losing
37 its ability to efficiently manage moisture. This effect indicates that the material starts saturating
38 as it can no longer retain water as efficiently. In the desorption phase, the material desorbs
39 moisture less efficiently demonstrating its inability to remove efficiently moisture. Moisture
40 retention will eventually lead to unwanted material decomposition.

41 'Group 3'
42



43 **Figure 9c.** Adsorption and desorption curves to show the categorisation of 3 samples within
44 'Group 3'.

Similar to 'Group 2', samples in this group adsorb moisture at a high rate for the initial 2 h of exposure. After that, moisture adsorption slows down significantly (see Figure 9c). On the contrary to the other groups exhibiting a peak after 8 h of exposure, – most of the samples within this group begin to desorb before the step change in RH. The adsorption/desorption curve of the samples in this group reaches saturation (a plateau within their adsorption/desorption curve) which corresponds to the inability of the material to exchange moisture.

Ideally in this study, materials are expected to steadily exchange moisture in a dynamic manner. Thus, samples of 'Group 1' are found to respond efficiently to differing hygrothermal environments, which is of primary importance when considering panels for indoor environmental comfort may render them ideal for. The shape of the adsorption/ desorption curves has been used to categorize the samples within Figure 8a and 8b (see Table 2).

Table 2. Categorisation of materials 7th Cycle and 10th Cycle through the adsorption/ desorption curve.

| Sample Name | Group Categorisation | |
|-------------|-----------------------|------------------------|
| | 7 th Cycle | 10 th Cycle |
| Wool 1 | 2 | 2 |
| Wool 2 | 2 | 2 |
| Wool 3 | 3 | 3 |
| Wool 4 | 2 | 2 |
| Hemp | 2 | 2 |
| WWB | 2 | 2 |
| SMR | 1 | 2 |
| Wood Fibre | 1 | 2 |
| Straw | 2 | 2 |
| ICB | 2 | 3 |
| PET | 2 | 3 |

Table 2 demonstrates that during the 7th cycle, this is not always the same group number as the final cycle categorisation. When differentiating between materials, the MBV may not be sufficient information to decipher one material from another. However, by being able to categorise a materials adsorption/desorption pattern would enable a more informed decision of the selection of a material to be made due to desired characteristics.

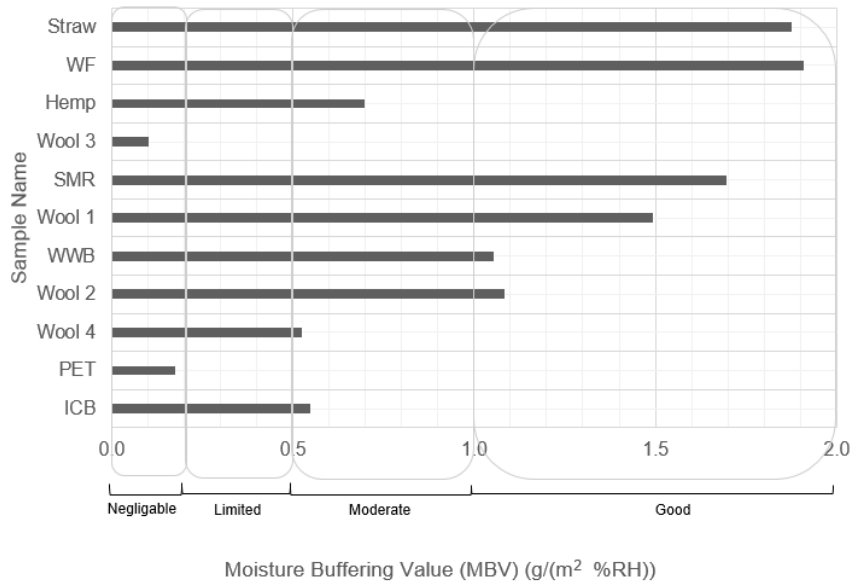


Figure 10. MBV of each material and its rating according to NORDTEST protocol, during the 10th cycle within climatic chamber.

Figure 10 demonstrates the classification of materials within this study employing a methodology outlined in (Rode et al, 2005). Out of 11 samples, 6 were categorised as 'good'; these bio-based materials were studied more extensively. Samples were put back into the chamber for 22 hygrothermal aging cycles as outlined in (Romano et al, 2018). The sample categorisation of samples from their final cycle is illustrated in Table 3.

Table 3. Material Properties.

| Material | 22nd Cycle Group Number | Mean MBV Value (g/ (m ² %RH)) | MBV Classification |
|----------|-------------------------|--|--------------------|
| Wool 1 | 1 | 1.88 | Good |
| Wool 2 | 1 | 1.23 | Good |
| WWB | 2 | Did Not Stabilise | - |
| SMR | 1 | 2.06 | Excellent |
| WF | 2 | 1.25 | Good |
| Straw | 2 | Did Not Stabilise | - |

Out of 6 bio-based materials that were within the chamber, only 4 materials stabilised and therefore have a MBV for comparison. SMR has the best category out of all the materials, whilst Wool 1, Wool 2 and WF have the same categorisation. When comparing their MBV, Wool 1 has the highest whilst Wool 2 and WF have very similar values. As there is only 0.02 g/(m² %RH) between them, this is not a sufficient differentiation between materials in order to select a preferential material. Because of this, the cycle group number must be considered in order to make a decision between Wool 2 and WF. As previously mentioned, Group 1 is the

109 most preferential group due to its efficient moisture exchange. After 22 cycles, Wool 2 has a
110 group classification of Group 1 rather than WF which is Group 2.

111 **3.3. Thermal Conductivity and SEM micrographs**

112

113 In order for an insulating material to be successful, it needs to buffer the way heat is exchanged
114 via radiation, convection and conduction (Callen, 1985). By measuring thermal conductivity,
115 this acts as an indication of the ability for a material to conduct heat. As aforementioned, the
116 thermal conductivity of the investigated samples was analysed. Figures 10a – 10v
117 demonstrates the thermal conductivity of each sample within a single 24 h hygrothermal aging
118 cycle, the samples surface morphology was investigated using SEM..

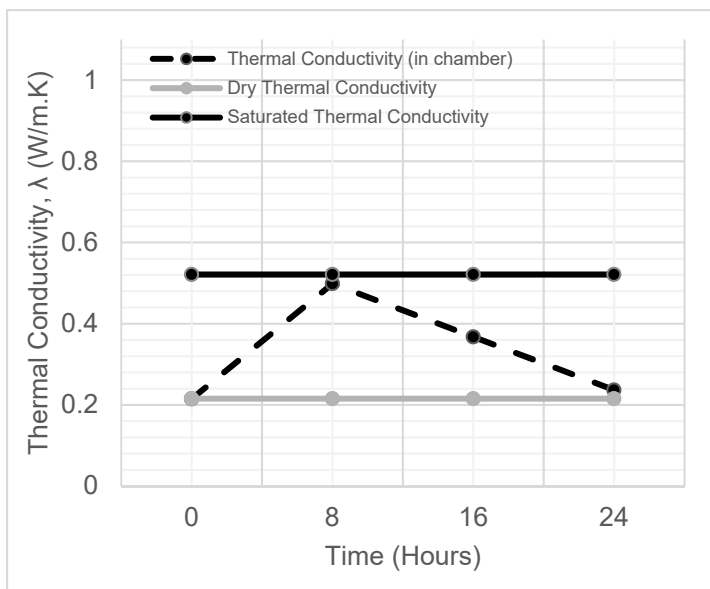


Figure 10a. PET - thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle

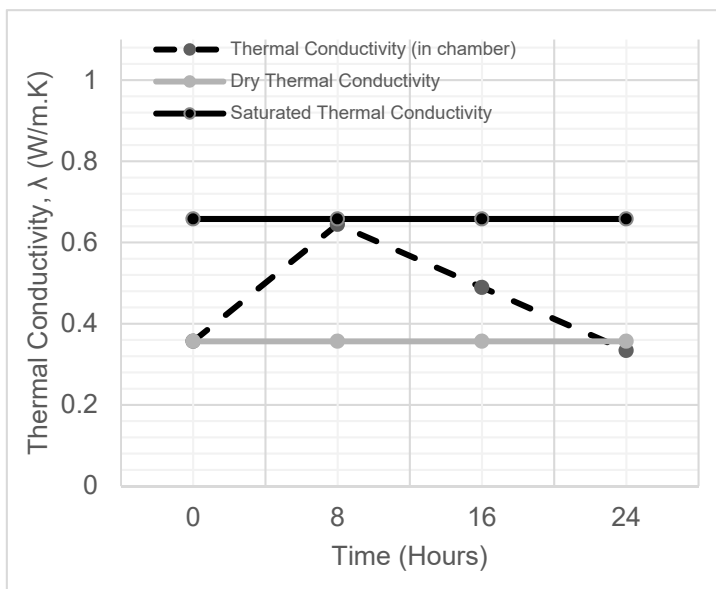


Figure 10c. Wool 2 - thermal conductivity of dry, saturated within a climatic chamber during a 24 hour cycle

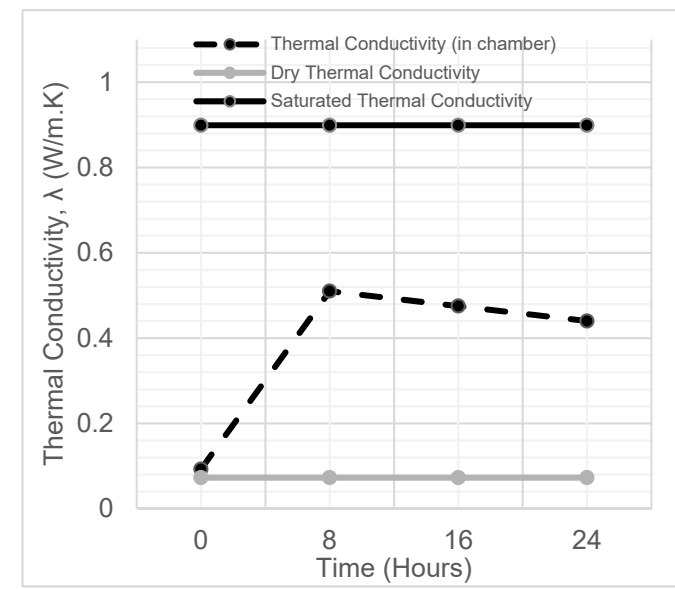


Figure 10e. WF - thermal conductivity of dry and saturated during a 24 hour cycle

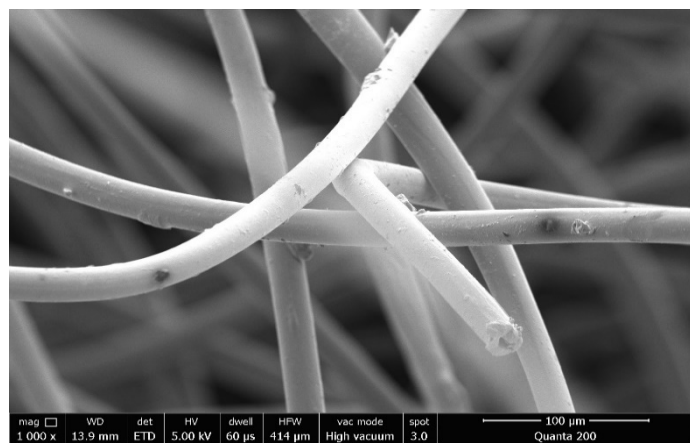


Figure 10b. SEM Image

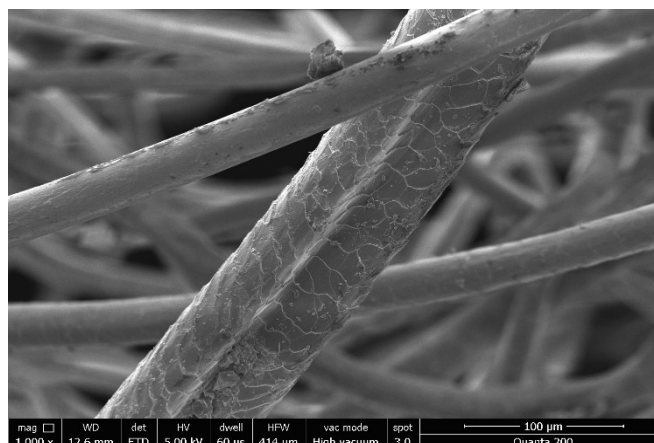


Figure 10d. SEM Image

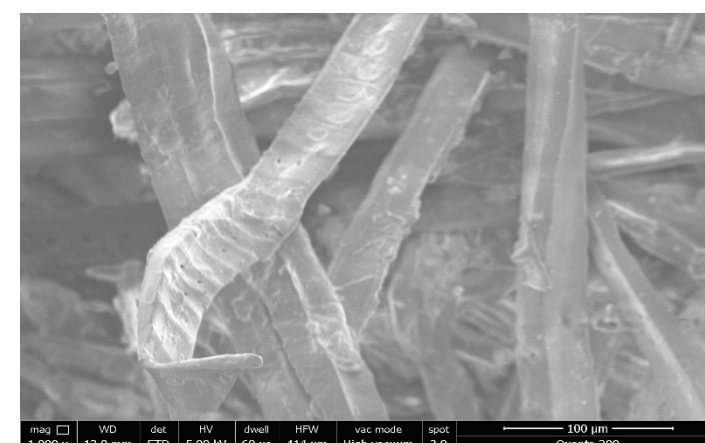


Figure 10f. SEM Image

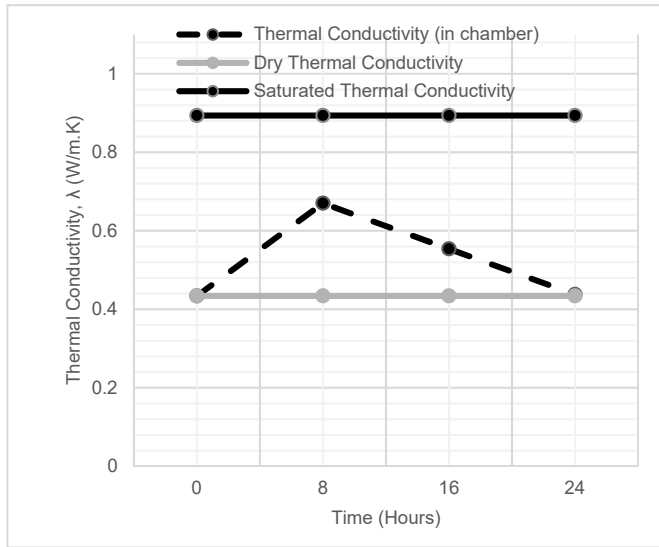


Figure 10g. Wool 3 - thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle.

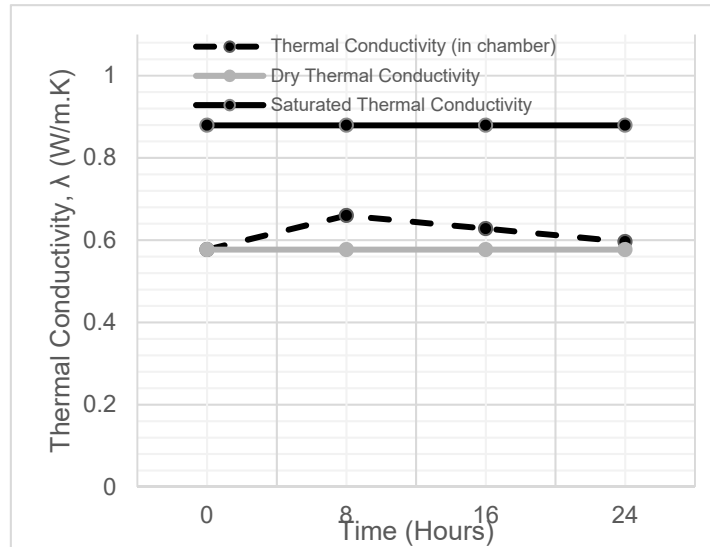


Figure 10i. Hemp - thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle.

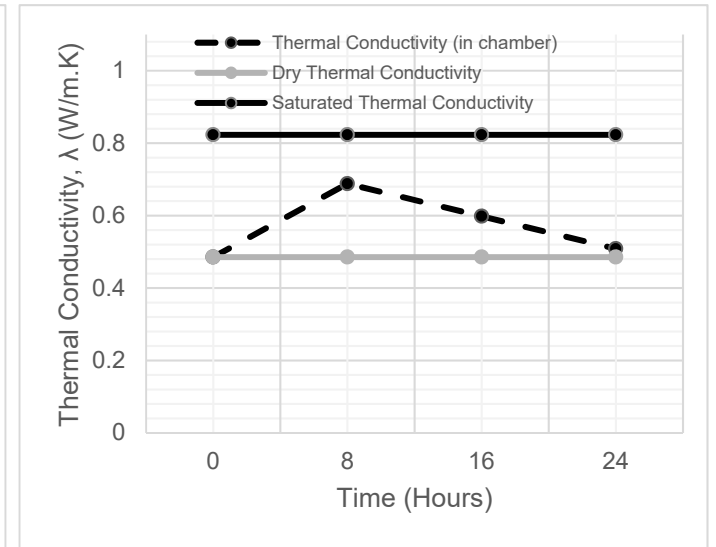


Figure 10k. SMR- thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle.

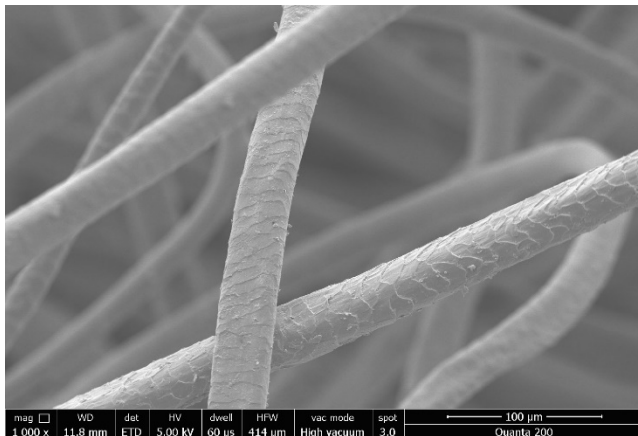


Figure 10h. SEM Image.

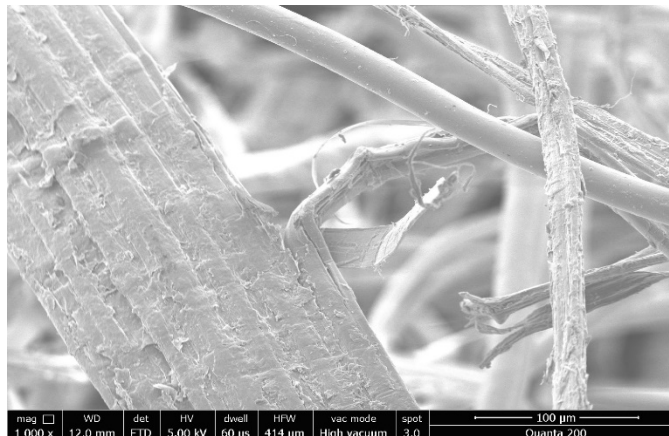


Figure 10j. SEM Image.

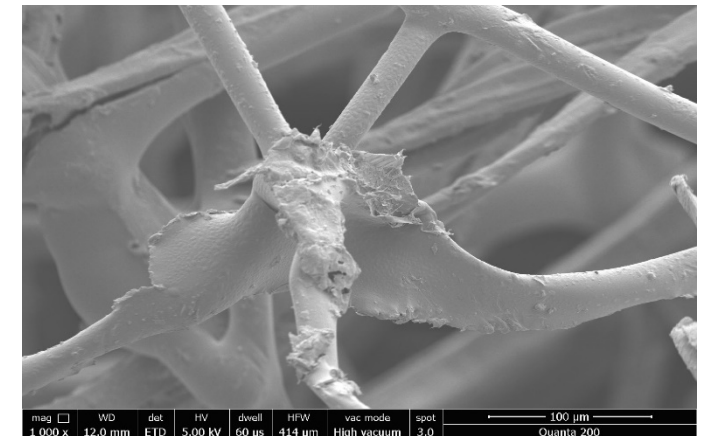


Figure 10l. SEM Image.

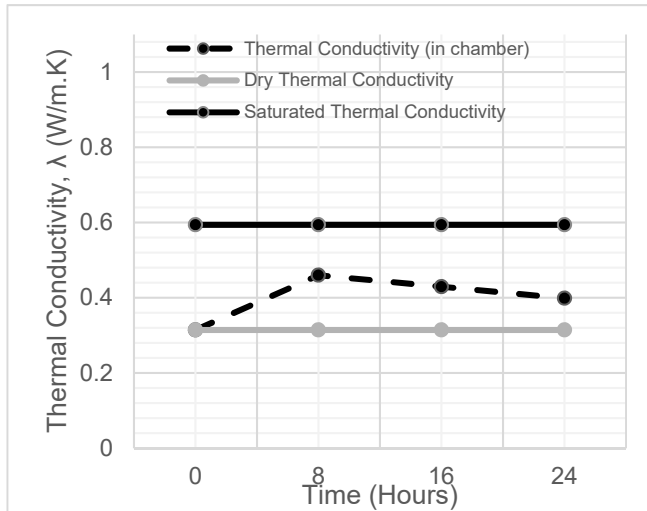


Figure 10m. Wool 1- thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle.

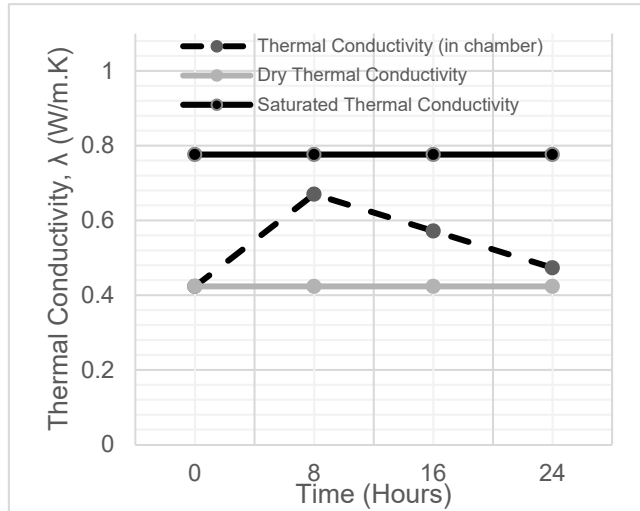


Figure 10o. Wool 4- thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle.

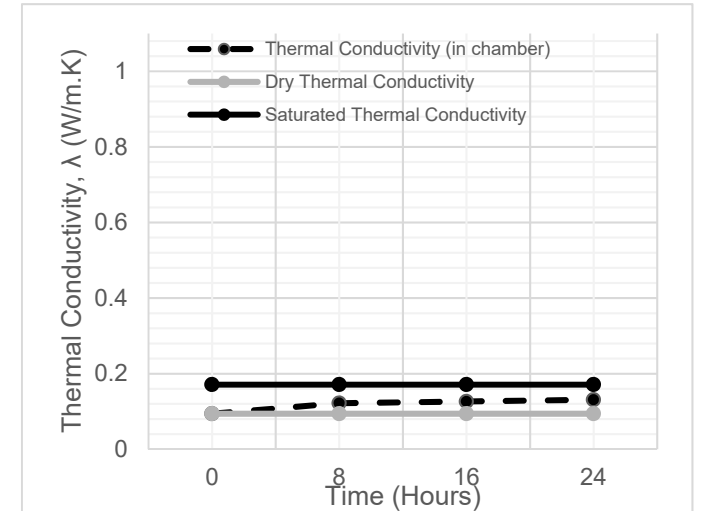


Figure 10q. ICB - thermal conductivity of dry, saturated within a climatic chamber during a 24-hour cycle.

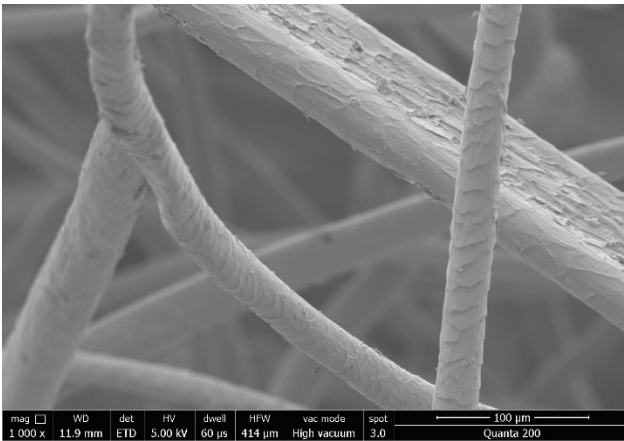


Figure 10n. SEM Image.

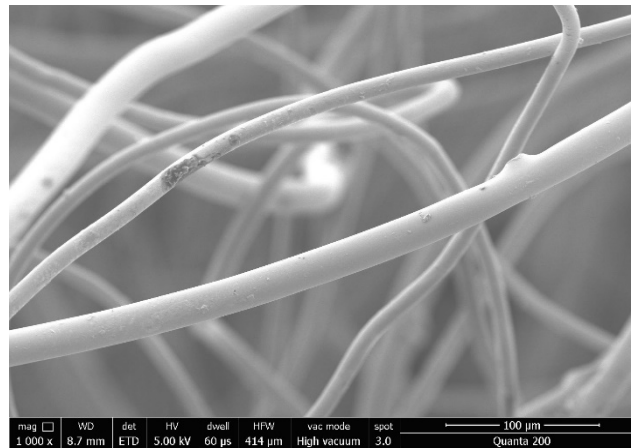


Figure 10p. SEM Image.

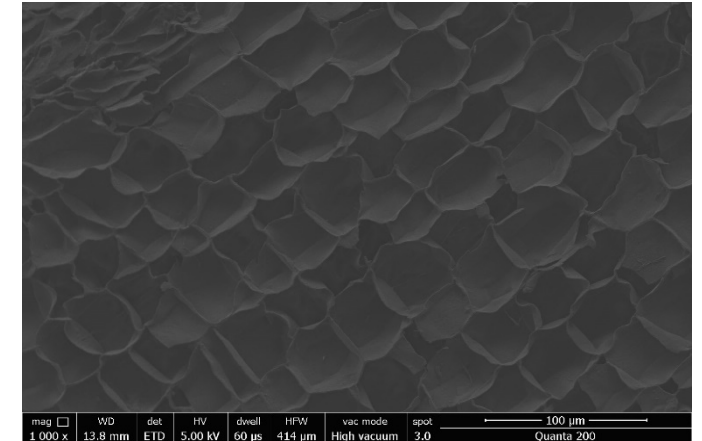


Figure 10r. SEM Image.

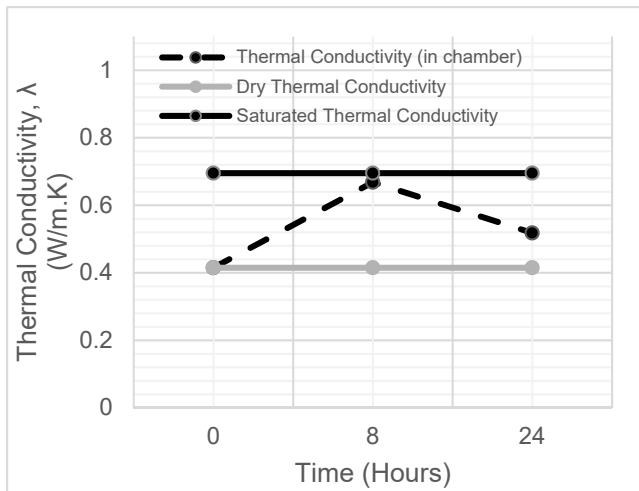


Figure 10s. WWB - thermal conductivity of dry, saturated within a climatic chamber during a 24 hour cycle.

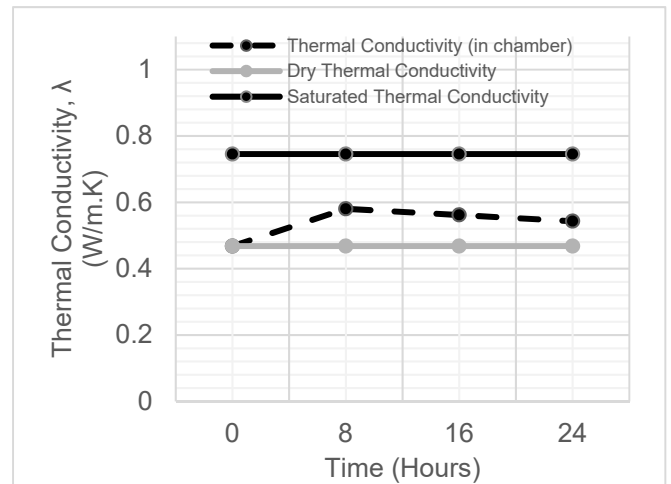


Figure 10u. Straw - thermal conductivity of dry, saturated within a climatic chamber during a 24 hour cycle.

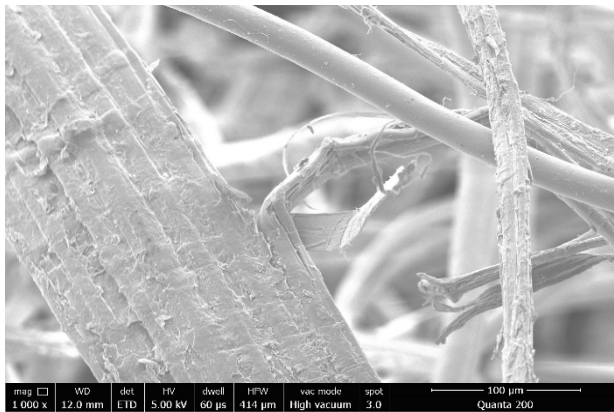


Figure 10t. SEM Image.

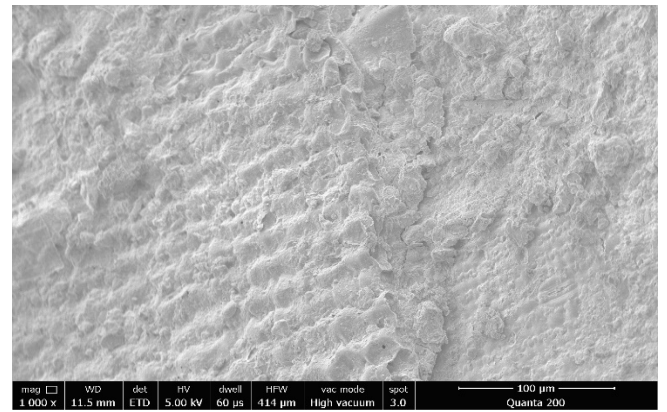


Figure 10v. SEM Image.

By comparing the thermal conductivity of dry and moisture-saturated samples one can conclude that the material loses its inherent insulating ability when saturated (Shea, Wall and Walker, 2013).

The thermal conductivity measurements obtained from the 11 studied materials can be classified into two different groups: those that exhibited identical and non-identical thermal conductivity values before and after hygrothermal cycling. In specifically, the samples that exhibited identical thermal conductivity values were: Wool 2, Wool 4, WFs, Hemp, PET and SMR. Whilst, the samples that revealed dissimilar thermal conductivity after exposure were: Wool 1, Wool 4, Straw, ICB and WWB.

The materials that revealed identical thermal conductivity values after exposure demonstrate a dynamic response to external hygrothermal changes. On the contrary, materials' inability to return their initial hygrothermal state (even if exposed for only hours and later remained at low RH for 16 h), retain water molecules within the structure, which reduces their ability to buffer moisture and function as an insulator. The latter case refers to non-hygroscopic materials (Zhang et al, 2017).

Further experimentation would be required to demonstrate the impact on measurements through further cycles and to understand whether the difference between 8-h peak and dry thermal conductivity readings. As the difference between the materials will either exaggerate throughout the experiment as the water molecules potentially become trapped and degrade /dissolve the bio-fibres or as the hygrothermal conditions change, the gap is at its largest and then lessens as the materials stabilises within the environment.

Figure 11a and 11b demonstrate the difference in thermal conductivity values as density varies within the two different states, that of dry and moisture-saturated samples. Within Figure 11a there appears to be no general trend, however within Figure 11b a linear trend demonstrates that thermal conductivity increases linearly with density increase. There are a few of exceptions to this, bio-based materials, exhibit an unexpected behaviour to hygrothermal exposure due to their nature (McGregor et al, 2016).

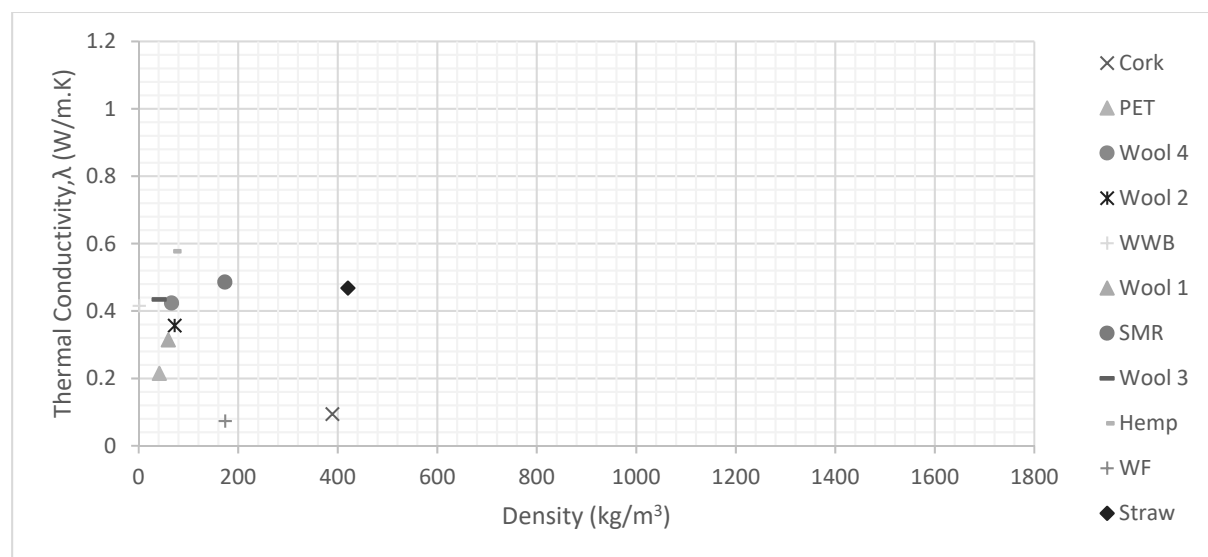


Figure 11a. A graph to show how thermal conductivity is affected by density whilst samples are dry (before starting MBV).

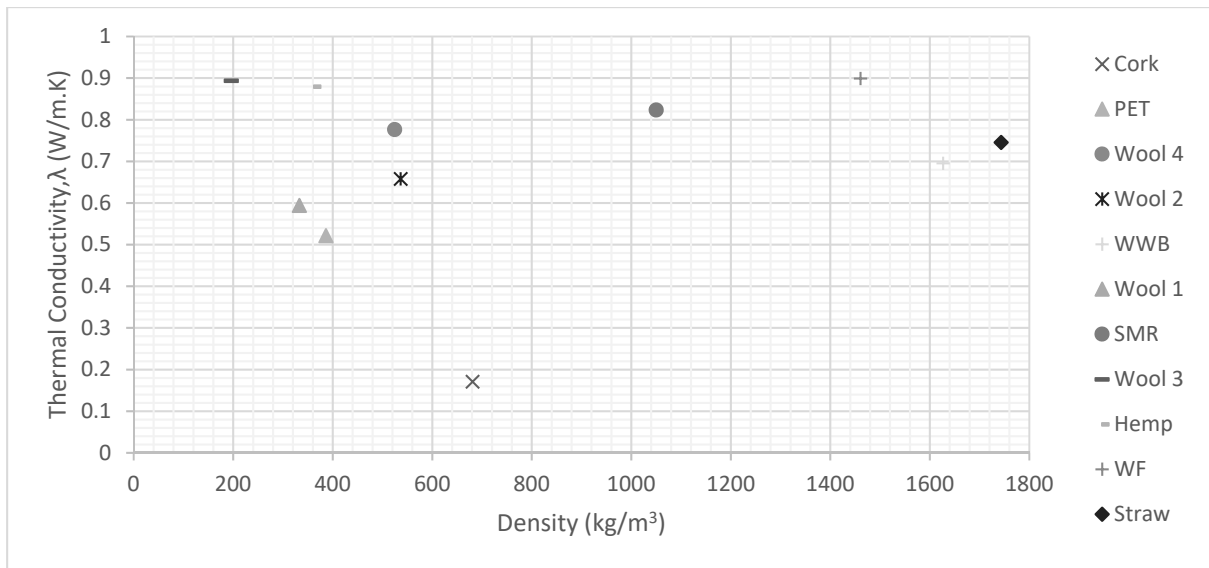


Figure 11b. A graph to show how thermal conductivity is affected by density whilst samples are saturated.

However, some of the outliers in Figure 11a – such as cork – could be attributed to the naturally occurring trapped air pockets and voids between ICB granules (see Figure 12). As presented within (Brás, Leal and Faria, 2013) not only this, but the microstructure of ICB is similar to a honeycomb providing further microscopic voids within the material (demonstrated in Figure 10r). Furthermore, having such a relatively low saturated thermal conductivity value exhibits the difficulty of water ingress to ICB cells in comparison to other materials (such as Wood Fibres). So, although this material has inherently low thermal conductivity value, its ability to be used within a ‘green panel’ is limited due to its low hygroscopicity.



Figure 12. ICB with naturally occurring air pockets and voids on the surface of the sample circled.

In comparison to other materials such as wool (see figures 10c, 10g, 10m and 10o), the voids are within the microstructure of the material rather than just between fibres. This contributes to how in the dry state, ICB can have a high density but equally low thermal conductivity. By comparing to previously conducted studies such as (Millogo et al, 2014) and (Bouguerra et al, 1998) the results within this papers align with them. When comparing Figures 11a and 11b it is clear that this data also correlates to the works of (Zach et al, 2012) where the higher the bulk density of a samples results in a more restricted air flow through and between the fibres of a material leading to less convection and therefore a higher thermal conductivity value.

When comparing the SEM images, it is evident that samples can be divided into 2 categories, those that demonstrate homogenous structure (see Figure 10r and 10v) and a more fibrous structure (in particular Figure 10p). A much more fibrous material indicates a multitude of plains for the material water molecules have a most 'free space' and voids to potentially fill and therefore more air pockets, equating to a lower thermal conductivity value.

In order for a material to be considered as an insulating material, it needs to exhibit a thermal conductivity value of less than 0.065 W/(m.K) (Pina dos Santos and Matias, 2006). When comparing the results of dry thermal conductivity values in Table 5 to the set value by Pina dos Santos (2006) none of the materials testes could be considered to have a thermally insulative material.

Table 5. Bio-fibres and dry thermal conductivity values in the dry state.

| Sample Name | Dry Thermal Conductivity W/(m.K) |
|-------------|----------------------------------|
| Wool 1 | 0.31 |
| Wool 2 | 0.36 |
| Wool 3 | 0.43 |
| Wool 4 | 0.42 |
| Hemp | 0.58 |
| WWB | 0.42 |
| SMR | 0.49 |
| WF | 0.47 |
| Straw | 0.47 |
| ICB | 0.22 |
| PET | 0.21 |

In comparison to data produced by (Thiebleson et al, 2017) where thermal conductivity ranges from 0.09 to 0.045 W/(m.K) for samples, the data of this study represent high thermal conductivity values which would render the studied materials as poor thermal insulators. However, when considering the thermal conductivity readings for both dry and moisture-saturated samples, it is important to consider that these materials have been through two sets of experiments within a climatic chamber in addition to being kept in a laboratory for week of inconsistent hygrothermal conditions. Furthermore, the results displayed in Table 5 are much higher than that of the thermal conductivity values as stated by manufacturers. Because of this, it is understood that samples can be affected by long exposure to hysteresis.

3.4. Temperature evolution during transient behaviour

Moisture buffering characteristics of bio-based materials renders them ideal materials for passive energy reduction management (Simonson, Salonvaara and Ojanen, 2004; Osanyintola and Simonson, 2006; Qin et al, 2011). A reduction in energy requirements can be attributed to the latent heat exchanges within the sample. Within this research paper, due to the continuous adsorption and desorption of water vapour within the bio-based samples there is an expectant latent heat exchange inherently to the structure of the bulk sample. Here, latent heat is attributed to the changing state (Hens, 2017) of water. During the phase change between water molecules and water vapour, latent heat is identified. During this phase change, a resultant change in temperature is expected, especially during the step change in RH values.

Figure 13 demonstrates the results from the thermocouples placed both on the surface and within the bio-based samples. It is evident from the figure that there has been no change in temperature throughout the experiment however; there is a distinct difference between the internal and external temperatures of the sample.

From a dynamic perspective, Figure 13 demonstrates that even after shifting the thermocouples the materials exhibit no changes in temperature per sample throughout the duration of the experiment. From a static perspective, there are some detected changes in temperature of samples when surface is compared to the temperature in the centre. This directly contradicts previous research that exhibits a change in temperature at a step change in RH (James et al, 2010; Holcroft and Shea, 2015). When comparing the duration of experiments, 'Run 1' lasts for 10 cycles of 24 hours whilst 'Run 2' lasts for 22 cycles of 24 hours. It is evident from both runs that there was a clear difference in temperature. The clear differentiation in temperature from within the sample to the surface and as there is no external heat source; the elevated temperature can only be attributed to latent heat.

Figure 14 displays that there is a multitude of characteristics between the different bio-fibres. Due to the temperature difference and due to their being no external heat source the temperature differential is due to latent heat. When considering the positive and negative values represented in Figure 15, these can be attributed to the latent heat vapourisation and condensation for which are not equal within all materials. SMR, Wool 1 and Wool 2 have equal temperature differential between Run 1 and Run 2 which demonstrates the ability for the material to stabilise within 22 cycles. However, WWB and Straw Run 2 demonstrates a high value in comparison to Run 1, despite Run 2 being 22 cycles is unable to stabilise within 10 cycles. Comparatively, WF exhibits a sustained negative temperature difference throughout both runs.

When considering the thermodynamics within the samples, generally (with exception to only WF) Run 1 demonstrates that the surface temperature is greater than the temperature at the centre of the sample (displayed as a positive value in Figure 14). Therefore, heat flow absorption into the sample is vapourised and as latent heat of vapourisation is an endothermic reaction and requires enthalpy to complete the process, the heat is absorbed by water to vapourise it. Run 2 is the converse, where the temperature at 15 mm from the surface is greater than that of the surface temperature so there is a heat flow release from the sample – (displayed as a negative value in Figure 14). WF demonstrates constant latent heat of condensation throughout both Run 1 and exaggerated particularly within Run 2. It is

hypothesised that due to the very nature of bio-based materials being intrinsically heterogeneous this could attribute that WF may not stabilise until after 22 cycles.

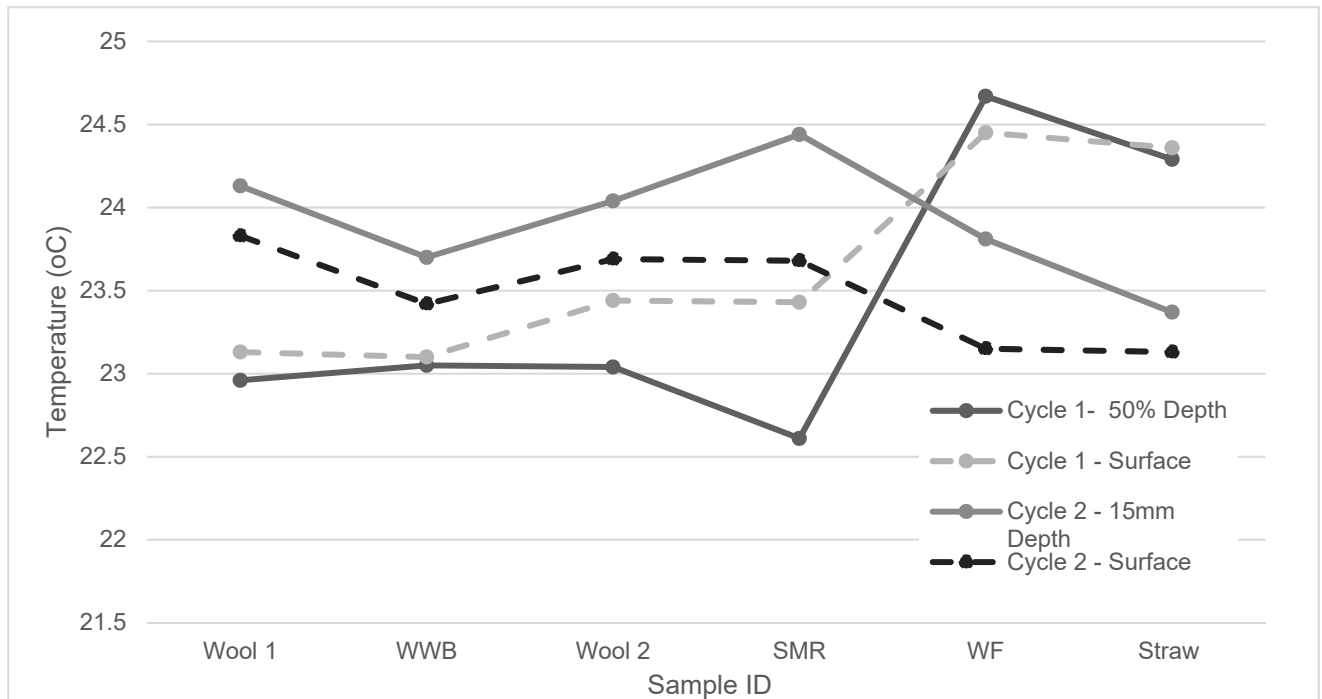
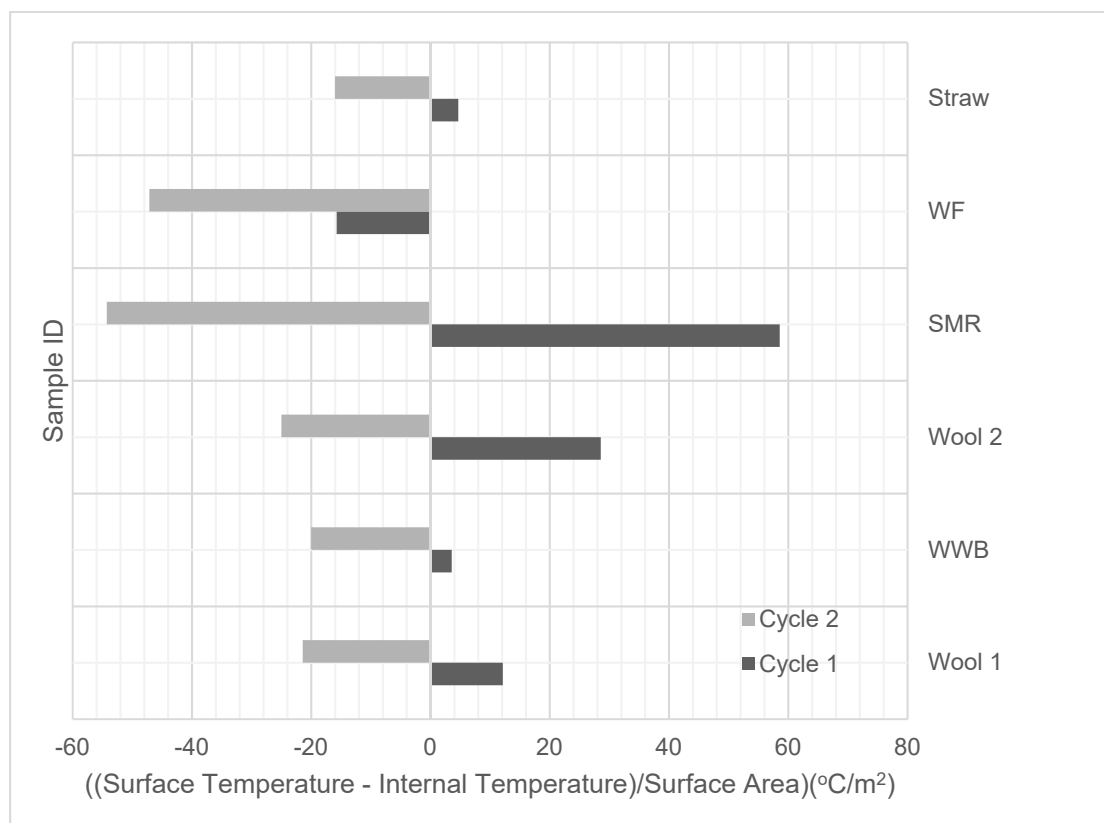


Figure 13. A graph to show temperature change on the surface and within samples throughout 2 experimental campaigns.

1



2

3

4

5 **Figure 14.** Temperature difference between surface and internal temperature per m² of bio-
6 based insulation sample (negative values: vapourisation; positive values: condensation).

4. Conclusion

Experimental testing on 10 bio-based and 1 recycled plastic insulation materials that are currently available on the market within the U.K. (4 different types of Wool insulation, Hemp, WWB, SMR, WF, Straw, ICB and PET), demonstrated which bio-based materials react the most effectively and efficiently to hygrothermal changes. They have been characterised by considering their dry and saturated density, thermal conductivity, MBV, steady state and transient temperatures and SEM images. Samples were exposed to relative humidity (RH) cycles of 75% and 53% for 8 and 16 hours (up to 22 cycles) to determine their Moisture Buffering Value (MBV). Dry/ wet thermal conductivity and density were monitored during exposure, whilst latent heat was determined through daily cycles. Scanning Electron Microscopy (SEM) was used to study the microstructure of the investigated material.

Results indicate that in order to select a material, simultaneously consideration of MBV and the shape of the final mass change graph is necessary to demonstrate adsorption/desorption characteristics of the sample.

It was found that only samples from SMR and Wool exhibited acceptable (MBV from 1 to 2 g/(m² %RH)) are one of the most promising as materials respond dynamic response to hygrothermal changes. Only half of the samples (Wool 2, Wool 4, WF, Hemp, PET and SMR, Wool 1, Wool 2) have the equivalent efficient moisture transfer to be able to desorb the same quantity of water. 'Green panels' made of bio-based materials are expected to passively control hygrothermal conditions and therefore thermal comfort within buildings. As hygrothermal conditions are an integral factor to this research, the MBV results are considered of primary importance. The highest MBV value in addition to lowest group number categorisation ensures the material is efficient and effective in terms of moisture exchange. On a microstructural level, the properties of each bio-based insulation material provide an explanation of their performance. After experimentation, two categories of samples were created. Preferable characteristics were revealed by the sample category that at the end of the cyclic exposure (whilst in the climatic chamber), the samples reverse back to their dry thermal conductivity values.

From the initial MBV, a total of 11 materials were tested, of which 6 that were categorised as 'Good', were tested further: Wool 1, Wool 2, WWB, SMR, WF and Straw. After these samples were then retested within the climatic chamber, the MBV categorisation results for Wool 1, Wool 2, SMR and WF were categorised as either 'Good' or 'Excellent' (see Table 3). Considering the shape of the final cycle, WF are categorised as 'Group 2' rather than 'Group 1', showing the other three samples as having superior characteristics. Thermal conductivity analysis demonstrates that on saturation, values rises of samples increases, where material dependant, samples cannot desorb water as efficiently as it is absorbed (as demonstrated in Figures 11a-11v).

From the SEM images taken of the bio-fibres, it is shown that the more fibrous samples have more potential air pockets, which would give an inherently lower thermal conductivity value. The SEM images demonstrate the fibrous nature of the samples and diversity of surface characteristics of the microstructure of bio-based materials.

This paper also demonstrates that latent heat can be detected in addition to also being further analysed and the categorisation of latent of vapourisation or condensation. When considering

the temperature evolution during transient behaviour, materials that can equally vapourise and condense (see Figure 15) have the most preferable properties - the bio-fibres that demonstrated this are Wool 1, Wool 2 and SMR.

Each of the different bio-fibres will have a slightly different mechanism for reacting to the hygrothermal environment due to the intrinsic properties and characteristics of bio-based materials. Future experimentation will demonstrate the combination of the bio-based fibres with other environmentally conscious materials in to boost their thermal properties. When considering all 10 bio-based materials and 1 recycled plastic samples and experimentation executed throughout this research paper, the samples for which will be taken forward for further development are: Wool 1, Wool 2 and SMR.

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