

NATURAL FIBRE INSULATION MATERIALS – THE IMPORTANCE OF HYGROSCOPICITY IN PROVIDING INDOOR CLIMATE CONTROL

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Abstract:

Interest in the use of natural fibres for insulation is increasing, due to the perceived environmental benefits of utilising materials in the built environment that contain sequestered atmospheric carbon. But the use of such materials in buildings presents both challenges and opportunities. For example, natural fibres absorb and desorb moisture as the relative humidity of the atmosphere changes. Adsorption is accompanied by the release of heat into the immediate environment and desorption results in the heat going into the material. This is known as the heat of sorption and it is additional to the heat flows associated with the evaporation or condensation of water (latent heat). The increasing use of such hygroscopic materials in buildings may have benefits regarding the ability to control the local environment passively, but before this can be analysed, it is necessary to determine the magnitude of the effect. The scientific literature provides details of how such measurements can be made, but when attempts were made to follow these, difficulties were encountered. A long study was undertaken to examine where the problems were and also to determine whether the assumptions made when determining heat of wetting were appropriate. The work presented in this paper represents the outcome of this investigation. Sorption isotherms were obtained for different natural fibres at various temperatures and the Clausius-Clapeyron equation applied to determine the heat of wetting. Data was obtained for Sitka spruce and for flax fibre. The applicability of this thermodynamic method is discussed in view of the fact that the presence of sorption hysteresis appears to imply that the material is not in thermodynamic equilibrium. This problem was dealt with in the past by employing inappropriate methods and these will be discussed. A model for hysteresis is presented and the implications of the use of thermodynamics to determine heat of sorption is discussed.

Keywords: *Natural fibres, insulation, water vapour, sorption*

1 Introduction

A sorption isotherm is obtained when a plant-based material is exposed to atmospheric relative humidity (RH) at constant temperature (hence the term isotherm). The normal procedure is to place a sample in a chamber of constant RH and allow sufficient time for

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the material to stabilise at the equilibrium moisture content (EMC). The sample is then weighed and removed to another chamber to stabilise at another RH, with the whole procedure taking place in a thermostatted environment. In the past, the procedure adopted was to control the RH by placing the samples in closed chambers containing various saturated salt solutions. Problems could be experienced if samples were removed from these chambers for weighing, although careful experimentation obviated such difficulties. It was established very early in the course of these studies that plant fibre and animal fibre materials exhibited a characteristic sigmoidal isotherm shape when the EMC was plotted against RH (as is shown in Figure 1). It was also found that if a sample was equilibrated at successively higher RH steps from an oven dry state, that the adsorption isotherm did not coincide with that of the desorption isotherm obtained when it was equilibrated at successively lower RH steps from a water saturated state, a phenomenon called hysteresis.

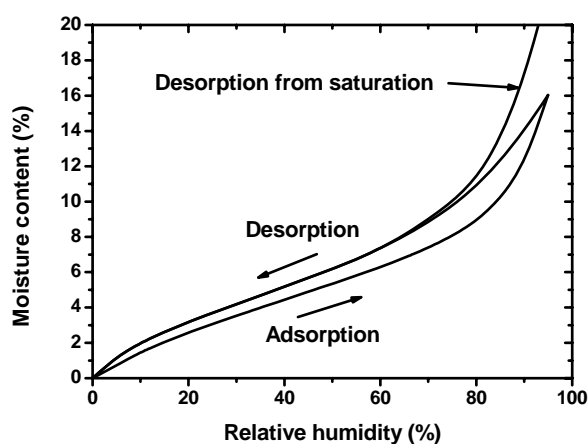


Figure 1: The sorption isotherm showing an adsorption isotherm a desorption isotherm from saturation (boundary curves) and the result of measuring the adsorption isotherm up to 95 % RH and then reducing the RH to zero again (the scanning curve)

If however, an adsorption isotherm is measured for an initially dry sample up to an RH of 95 % and then the RH is reduced again a new curve is obtained (called a scanning curve) which approaches and then joins the boundary desorption isotherm curve. The original interpretation for the property of hysteresis was that when a sample was dried from a water-saturated state to dryness, irreversible hydrogen bonds were formed that could not be broken again and therefore were no longer available to water molecules (hornification). It was also realised that since the adsorption and desorption boundary curves of the isotherm did not follow the same pathway, then the sample could not be considered to be in true equilibrium with its environment and that as a consequence thermodynamic principles could not be applied. In order to deal with these issues, Stamm and Loughborough (1934) introduced the concept of an 'oscillating vapor pressure desorption curve' in which it was supposed that a true (or at least pseudo-equilibrium) was established. This then allowed for determination of the heat of sorption by determining the sorption isotherms at different temperatures and applying the Clausius-Clapeyron equation (Equation 1). The oscillating isotherm was obtained by measuring the moisture contents of relatively large wood samples at various RH and temperature settings. Since the oscillating isotherm was found to fall between the boundary curves delineated by the adsorption and desorption isotherms, it was considered that this was a good approximation to the 'true' equilibrium isotherm. Since that time there have been many

studies where the Clausius-Clapeyron equation has been used to determine heat of sorption and various text books describe the methodology to be adopted. Thus it was the original intention of the research project described herein to use this technique to calculate the heat of sorption in order to evaluate the magnitude of the effect. Life cycle assessment studies do not take account of the heat of sorption phenomenon when describing the use phase of natural fibre insulation materials. It remained a possibility that heat flow into and out of a mass of fibres associated with desorption and adsorption processes might have an important effect on the life-cycle impacts when considered throughout a buildings lifetime.

2 Determination of the Sorption Isotherms

Samples of plant fibre material were exposed in a dynamic vapour sorption (DVS) apparatus (Surface Measurement Systems Ltd, London UK). A typical isotherm run started at zero percent RH and the RH was then increased in a number of 5 % or 10 % RH increments until an RH of 95 % was reached, then the desorption part of the isotherm loop was determined. Full experimental details have been given in Hill et al. (2009). A range of plant fibres were studied in this experiment, although the effect of different temperatures was only studied for flax and Sitka spruce. Flax was of interest, since the study was to be applied to a commercial product which had flax as a major component. Sitka spruce was investigated in order to provide comparison with literature values. The results of a typical isotherm experiment are given in Figure 2.

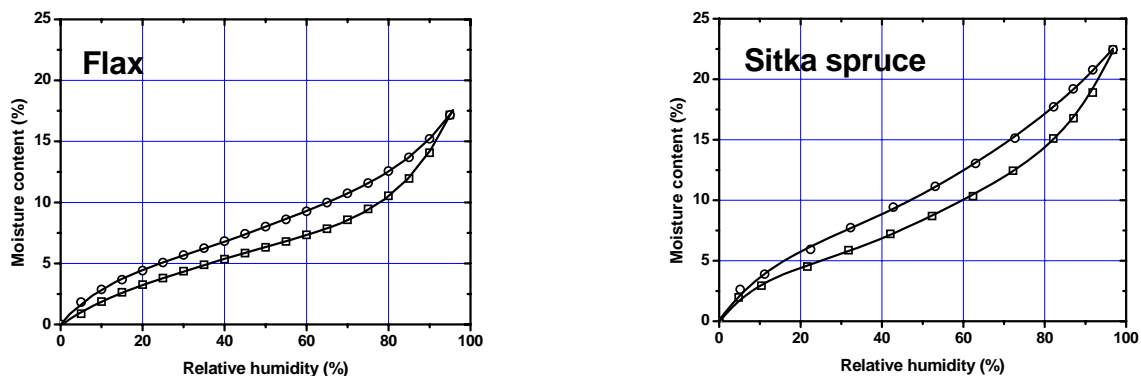


Figure 2: Sorption isotherms of flax and Sitka spruce at 25 °C

These plots are typical of the two species studied in that Sitka spruce exhibits higher moisture contents over the hygroscopic range and the area bound by the hysteresis loop is larger than with flax. By examining a large range of plant fibres, it has been found that fibres with a higher lignin content also exhibit larger hysteresis. This is consistent with the expected behaviour for sorption into solids below the glass transition temperature (Hill et al. 2009).

3 Temperature Dependence of the Isotherms

According to the scientific literature, the sorption isotherm exhibits the temperature relationship shown in Figure 3 (adapted from Skaar 1972). The sorption isotherm moves to the RHS as the measurement temperature is increased, so at any given RH the MC of the sample is lower. In order to determine the heat of sorption it is necessary to determine the different RH values at various temperatures where the MC is constant (isosteres) as

shown in Figure 3. If the isotherm is measured at two (preferably widely spaced) temperatures, the Clausius-Clapeyron equation (Equation 1) can then be used to determine the heat of sorption.

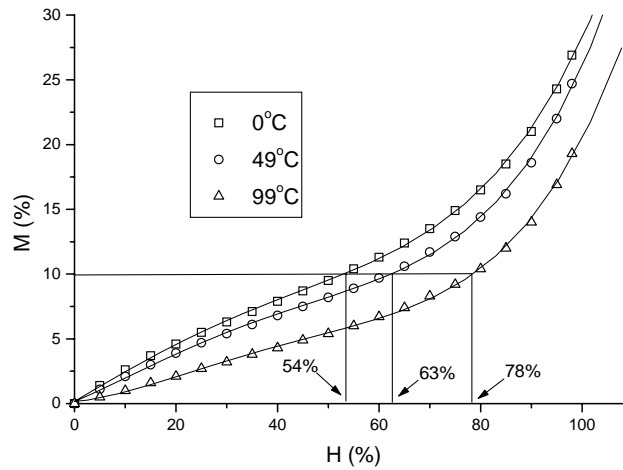


Figure 3: Sorption isotherms plotted for different temperatures (data taken from the Wood Handbook, USDA 1974). The procedure requires the determination of RH (H) values at different temperatures for constant MC (M)

$$Q_v = -1.60172[\log(p_1) - \log(p_2)] / [(1/T_1) - (1/T_2)] \quad (1)$$

Where Q_v is the differential heat of wetting and p_1 and p_2 are the vapour pressures of water at temperatures T_1 and T_2 respectively.

It was at this point that difficulties were encountered. Determination of the adsorption isotherms at different temperatures failed to show any differences; the plots were identical (Figure 4). This was found for all fibres studied.

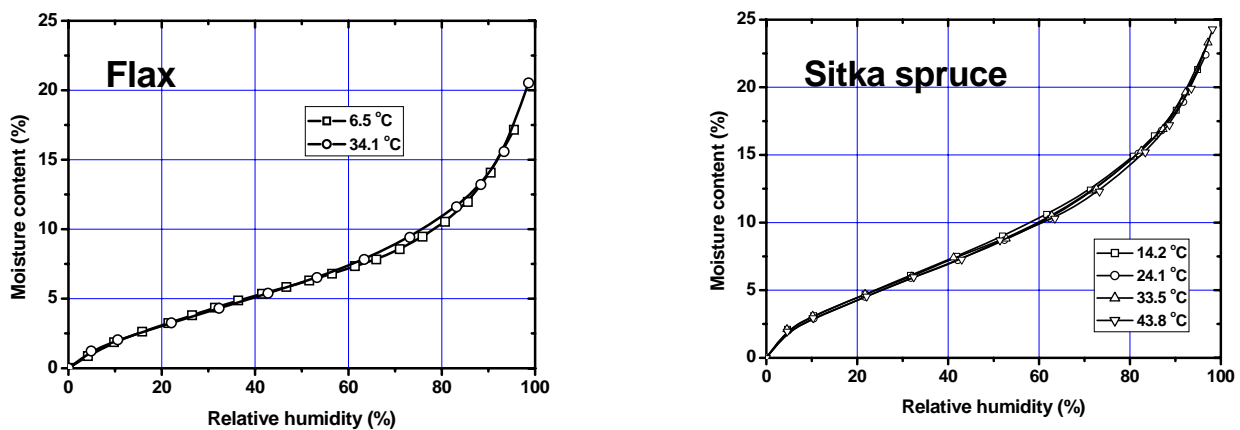


Figure 3: Adsorption isotherms for flax and Sitka spruce plotted for different temperatures

This result was not expected and has not been reported in the literature previously. However, when the desorption isotherms at different temperatures were plotted, it was found that the predicted behaviour did occur (Figure 4). As previously noted in this paper, Stamm and Loughborough (1934) did not determine adsorption nor desorption isotherms

in their work. Over the years their work has been used as the basis for many thermodynamic studies and the data that they obtained appears to be that subsequently quoted in the Wood Handbook (USDA 1974) and used many times when various thermodynamic analyses of isotherms are reported. But, it is important to note that this is not a true isotherm. The original graph from the Stamm and Loughborough (1934) paper is reproduced in Figure 5. This clearly shows the ‘oscillating vapor pressure desorption’ curve falling between the adsorption and desorption curves.

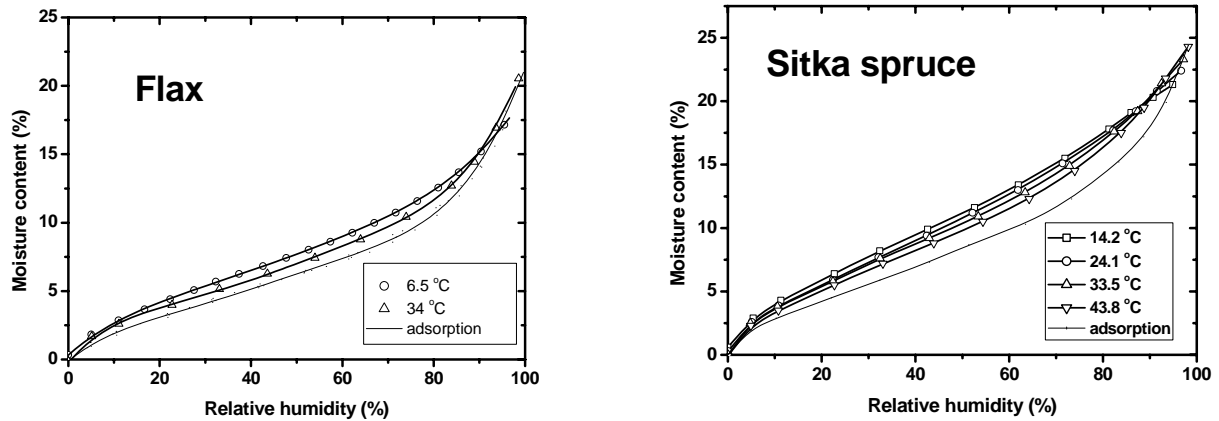


Figure 4: Desorption isotherms for flax and Sitka spruce plotted for different temperatures the average adsorption line is shown without data points for comparison

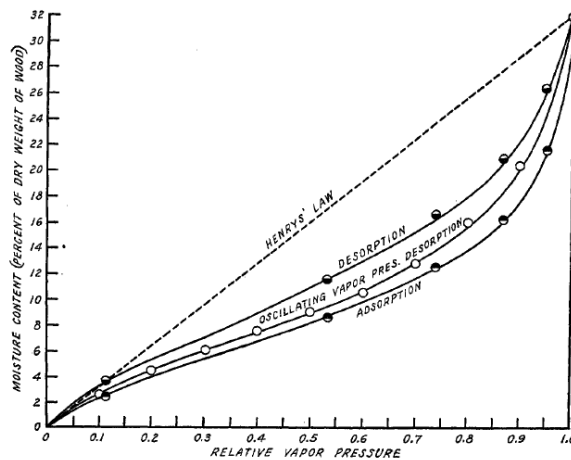


Figure 5: Original graph reproduced from Stamm and Loughborough (1934) showing the ‘oscillating vapour pressure desorption isotherm’

4 Origin of Hysteresis

The results from this study show that the adsorption isotherm curve is static and independent of temperature whereas the desorption curve changes in the predicted manner as the isotherm temperature is increased. The net result of this is that the area bounded by the hysteresis loop decreases as the temperature is increased (Figure 4). This behaviour is what would be predicted for sorption processes on glassy solids (Hill et al. 2009). As the glass transition temperature (T_g) is approached, the size of the hysteresis loop decreases and becomes zero at T_g . Measurements of the sorption isotherms were not made at temperatures sufficiently close to the expected T_g of water-plasticized lignin

(about 100 °C), but projections of the hysteresis values fall to zero at 100 °C, which is entirely consistent with the model presented herein.

5 Is the use of the Clausius-Clapeyron Equation Valid?

There has been a long-running debate in the literature as to whether it is appropriate to use thermodynamic analyses such as the Clausius-Clapeyron equation when studying sorption phenomena. Thermodynamics can only be used for reversible systems, meaning that the paths of the adsorption and desorption curves should be identical, which is not the case. It can only be concluded that the use of thermodynamics in this respect is at best open to question and more probably invalid. Thus, the only way that heat of wetting phenomena can be reliably studied is through the use of calorimetric techniques to give the integral heat of wetting. The use of hygroscopic building materials such as natural fibres having a high surface volume ratio creates new possibilities for the exploitation of properties such as the heat of sorption in designing passive climate control systems as well as providing insulation. If properties such as these are to be used then they need to be understood. One approach is to build test structures and monitor their performance. However, it is also very important that we can understand these properties at a fundamental level so that this knowledge can be incorporated at the design stage, thereby saving unnecessary expense and expenditure of time. If the thermodynamic approach is questionable, as appears to be the case, then it is important to determine the heat of sorption using calorimetric methods in conjunction with water vapour sorption. Such studies are lacking at the present time.

6 References

- HILL, C.A.S., NORTON, A.J., NEWMAN, G., 2009. The water vapor sorption behavior of natural fibers. *Journal of Applied Polymer Science*, 112(3), pp. 1524-1537.
- SKAAR, C., 1972. *Water in wood*. Syracuse University Press, New York.
- STAMM, A.J., LOUGHBOROUGH, W.K., 1934. Thermodynamics of the swelling of wood. *Journal of Physical Chemistry*, 39(), pp. 121-132.
- UNITED STATES DEPARTMENT OF AGRICULTURE, 1974. *Handbook No. 72 – Wood as an Engineering Material*. US Government Printing Office Washington DC, USA.