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THE IN-PLANE AND OUT-OF-PLANE HYGROEXPANSIONAL PROPERTIES OF PAPER

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ABSTRACT

This paper describes some studies of the effects of drying restraints and sheet density on the in-plane and out-of-plane hygroexpansivity of paper. It is shown that drying restraints in the RH range, i.e. between 20% and 0% moisture content, greatly influence hygroexpansivity, with in-plane hygroexpansivity being lower the lower the RH to which the paper has been dried under restraint. For sheets of high density, the volume expansivity is not affected by the drying restraint, and the reduction in in-plane hygroexpansivity is compensated by an increase in out-of-plane hygroexpansivity.

For freely dried sheets both the in-plane and the out-of-plane hygroexpansivity increase with increasing density. The volume hygroexpansivity at various densities is similar to that for wood of the same density. For sheets dried under restraint the density has only a slight influence on the in-plane hygroexpansivity. The out-of-plane hygroexpansivity is higher than for freely dried sheets but includes changes which are probably irreversible, particularly at low densities.

INTRODUCTION

For many applications, the hygroexpansivity of paper is an important property which affects for example printing operations, the runnability of fast packaging machines and the runnability of computer papers. An understanding of the fundamental parameters affecting the hygroexpansivity is essential in order to develop more dimensionally stable papers for these applications. The factors of most importance are the degree of beating and the drying shrinkage of the pulp, the sheet anisotropy and the drying restrictions imposed upon the paper during drying (1). The hygroexpansion of the paper is also governed by the swelling capability of the wood polymers in the fibre. Hygroexpansion is therefore dependent on the physical components of the constituent fibers as well as the configuration of these components in the fibre and in the fibrous network.

In this study, the effects of some aspects of paper structure on the in-plane and out-of-plane hygroexpansivity are demonstrated. Handsheets were subjected to different drying conditions, and the effect of density on both freely dried sheets and sheets dried under restraint was examined. The relationship between in-plane and out-of-plane hygroexpansivity is also discussed.

MOISTURE SORPTION

The hygroexpansion of paper is a result of the hygroscopic nature of its constituent wood polymers. The moisture content of the fibre depends on both chemical and physical factors. At the fibre saturation point or above, the moisture content, as for instance given by the Water Retention Value (WRV), is dependent on the void volume and the internal and external surface area of the fibre, the sorptive components of the fibre and the swelling restrictions that are applied by the fibre structure. At lower moisture contents, the sorption is mainly determined by the number of accessible hydroxyl groups, with other structural factors playing a minor role.

The effect of drying conditions on the moisture sorption is shown in Fig.1, which compares the sorption isotherm of a freely dried sheet with that of a sheet dried under restraint. No major difference in moisture sorption can be noticed between these two sheets, even though their hygroexpansional properties are quite different, as will be shown later. In the following it is considered that all the tested papers have identical sorption isotherms, regardless of drying conditions.

In some cases it has been reported that at high humidities, i.e. at 90% RH, the equilibrium moisture content of a high density paper material is lower than for the corresponding material at a lower sheet density (2). However, no such effects have been reported for paper at conditions of lower RH. In this study, measurements are only reported up to 65% RH, so the effect of density on the moisture content of the paper is considered marginal and is here neglected.

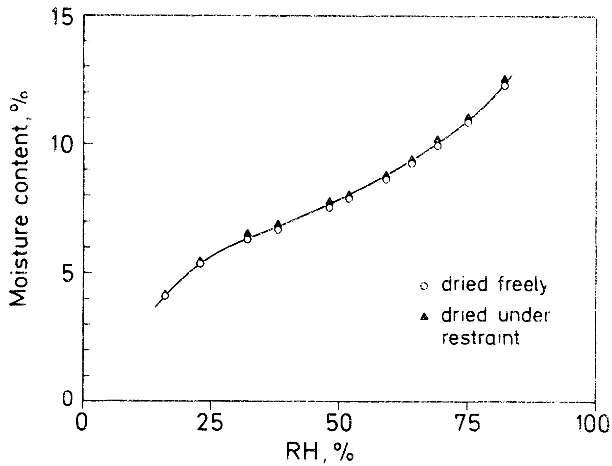

Fig 1—Moisture sorption isotherm at 23°C for sheets dried either freely or under restraint

OBJECTIVE OF THE INVESTIGATION

The objective of this investigation is to investigate the effects of sheet density and drying strategies on the in-plane and the out-of-plane hygroexpansivity of paper. For this purpose, sheets were wet-pressed to give various densities and then dried under restraint to different final dry solids contents. This was achieved by drying to equilibrium at different relative humidities, as shown in the drying strategy scheme (Fig.2). Prior to the hygroexpansion measurements, the sheets were conditioned at 16% RH in order to diminish moisture hysteresis. The hygroexpansivity was then determined between 25% RH and 65% RH. In this range, a linear relationship exists between hygroexpansion and moisture content, regardless of drying strategy.

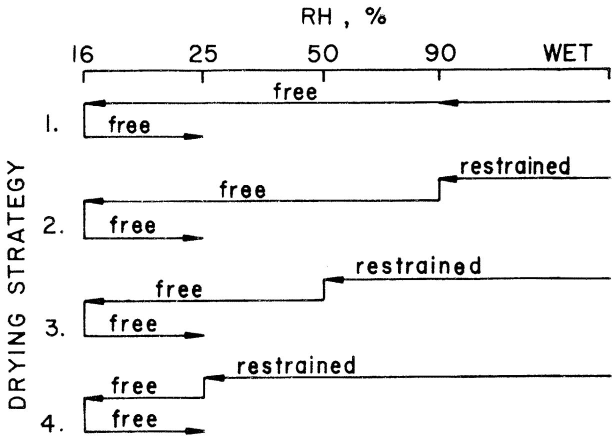

Fig 2—Drying strategy schemes for varying degree of restraint

In common with other paper properties, hygroexpansion exhibits an hysteresis effect versus relative humidity. However, for freely dried sheets the dimensions are reversibly determined only by its moisture content (1). For sheets dried under restraint, a release of dried-in stresses during moisture cycling is a permanent shrinkage of the sheet (1,3). In this investigation, hygroexpansion was only followed over the first adsorption isotherm up to 65% RH. Therefore the recorded hygroexpansion for papers dried under restraint may be partly irreversible, since a release of structural restraints may take place when the humidity is increased. The effects of moisture cycling are not within the scope of this paper.

RESULTS

Effects of Drying Restraints

The effect of varying the degree of drying restraint on the in-plane hygroexpansivity of isotropic sheets is exemplified in Fig.3. For sheets which have been dried under restraint, the in-plane hygroexpansivity decreases as the final RH at which the paper is released from restraint decreases. Furthermore, the difference in hygroexpansivity between sheets that have been subjected to drying restraints to 90% RH and 25% RH is of the same order of magnitude as between sheets that have been dried freely and those dried under restraint to 90% RH.

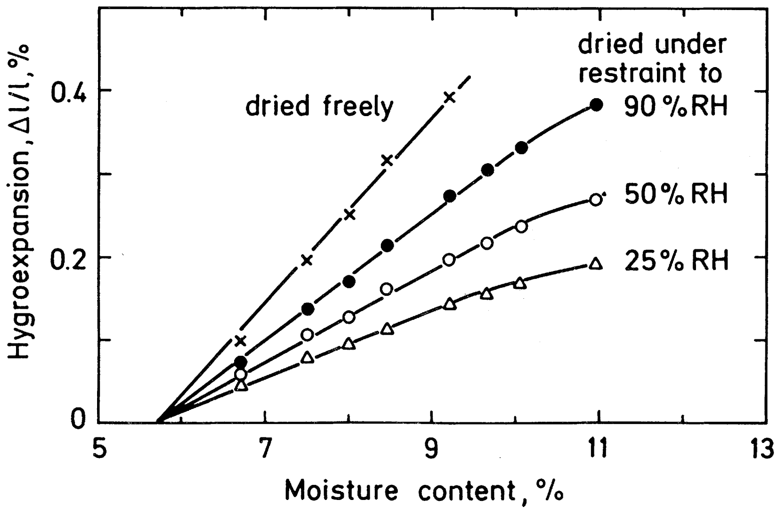

Fig 3—In-plane hygroexpansion (relative length increase Δℓ / ℓ) versus moisture content for isotropic sheets dried either freely or under restraint down to different RH-levels.

For oriented sheets, the in-plane hygroexpansivity is higher in the CD than in the MD (1), (Fig.4). The effect of the final relative humidity to which the sheet has been dried under restraint, both in the MD and the CD, is similar to that shown for isotropic sheets in Fig.3.

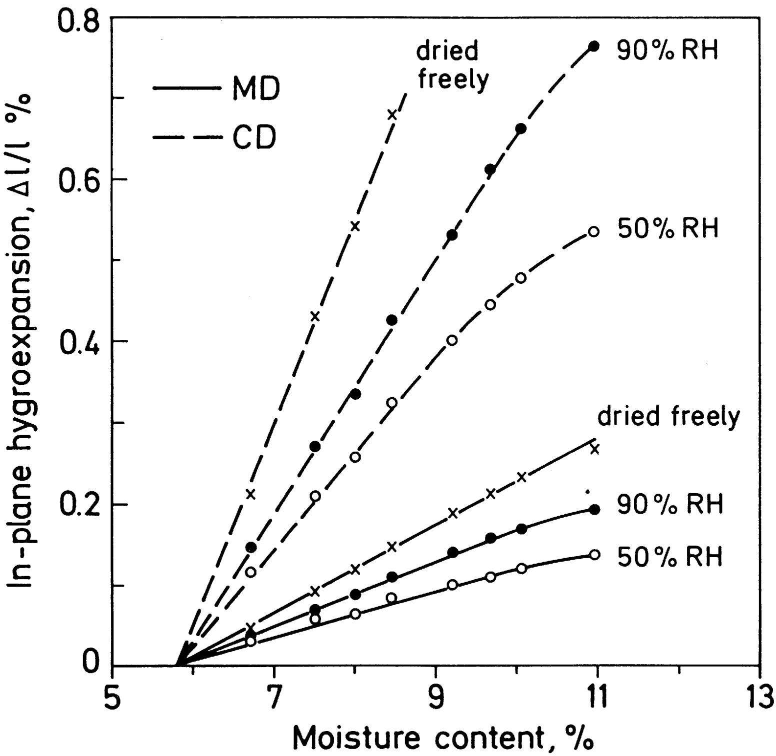

Fig 4—In-plane hygroexpansion versus moisture content for oriented sheets dried either freely or under restraint down to different RH-levels.

From Fig. 5 it is evident that a decrease in the in-plane hygroexpansivity caused by drying restraint is compensated by an increase in the out-of-plane hygroexpansivity. In this case, an inverse relationship exists between the in-plane and the out-of-plane hygroexpansivity. It should also be noted that the out-of-plane hygroexpansivity is an order to magnitude higher than the in-plane hygroexpansivity.

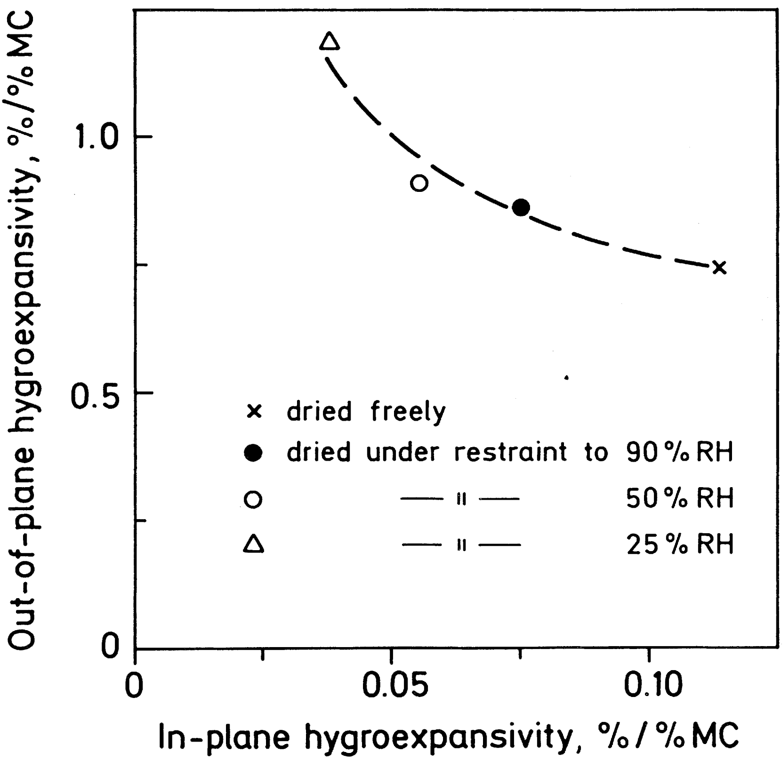

Fig 5—Out-of-plane hygroexpansivity versus in-plane hygroexpansivity for isotropic sheets dried either freely or under restraint down to different RH-levels. The hydroexpansivity is given in % change in length per % increase in moisture content (MC)

Effects of Density

The influence of sheet density on the in-plane hygroexpansivity of isotropic sheets is illustrated in Fig.6. For the freely dried sheets (drying strategy 1. in Fig.2) the hygroexpansivity increases with increased density. Conversely, the in-plane hygroexpansivity of the sheets dried under restraint (drying strategy 3. in Fig.2) slightly decreases with increasing density.

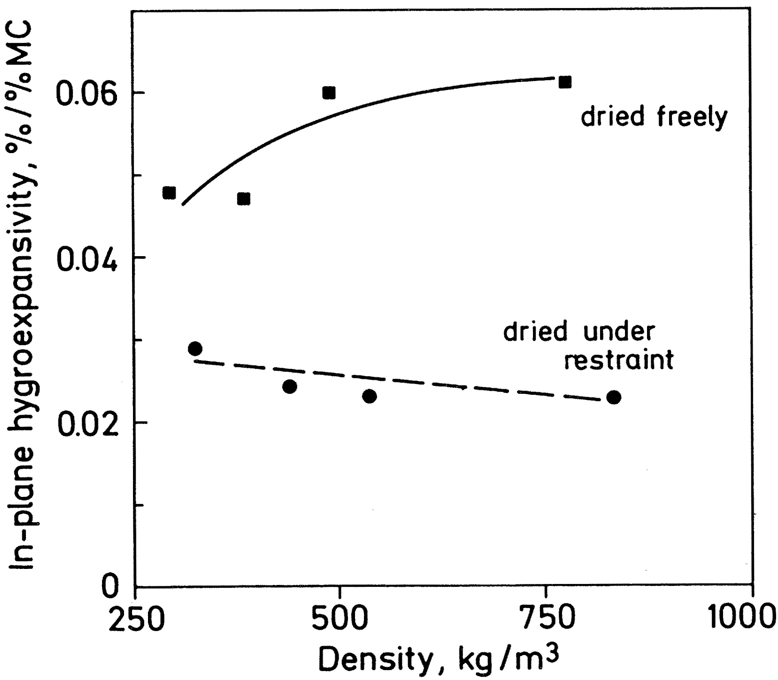

Fig 6—In-plane hydroexpansivity versus density for isotropic sheets dried either freely or under restraint to 50% RH. The various densities were achieved by different wet-pressing pressures.

Fig.7 shows that the in-plane hygroexpansivity of freely dried oriented sheets increases with increasing density only in the CD. The hygroexpansivity seems to the unaffected by density in the MD. For the sheets dried under restraint only a slight decrease with increasing density is noticed in the MD, while the hygroexpansivity in the CD is constant.

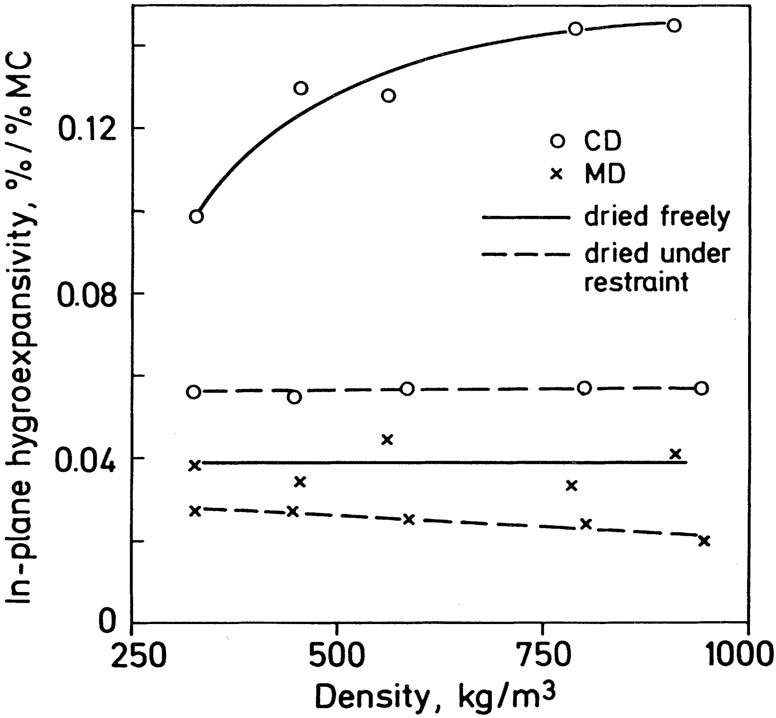

Fig 7—In-plane hydroexpansivity versus density for oriented sheets dried either freely or under restraint to 50% RH. The various densities were achieved by different wet-pressing pressures.

The effect of density on the out-of-plane hygroexpansivity is shown in Fig. 8. For sheets dried freely, the hygroexpansion increases appreciably with increasing density. For sheets dried under restraint, the hygroexpansivity slightly decreases with increasing density. It is always higher than the corresponding free-dried value, but the values are tending to converge at higher density.

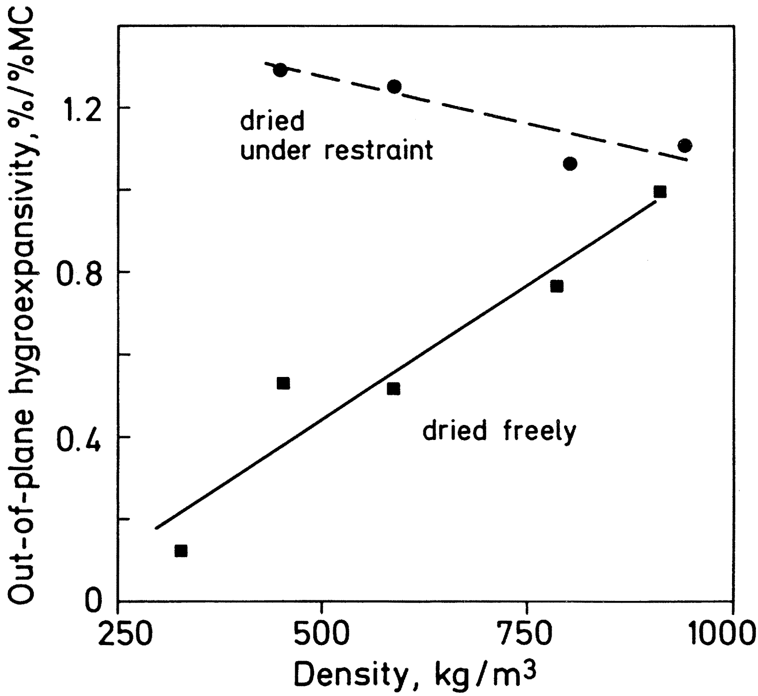

Fig 8—Out-of-plane hydroexpansivity versus density for oriented sheets dried either freely or under restraint to 50% RH. The various densities were achieved by different wet-pressing pressures.

In Fig.9 the out-of-plane hygroexpansion is plotted versus the in-plane hygroexpansion (geometric mean value) for oriented sheets of different density. When the density is changed for a sheet subjected to a given drying procedure, it seems that a linear relationship exists between the in-plane and out-of-plane hygroexpansivity.

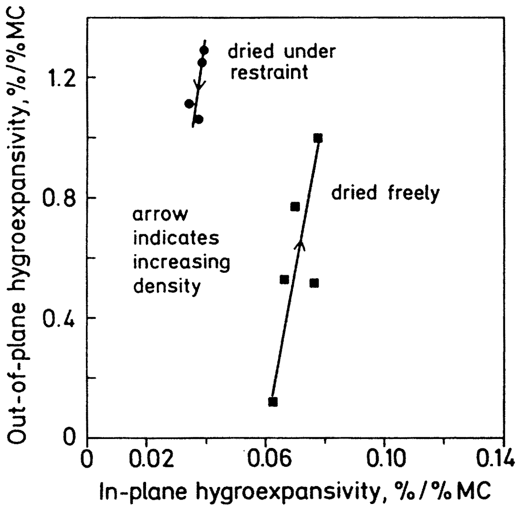

Fig 9—Out-of-plane hydroexpansivity versus in-plane hydroexpansivity (geometric mean value) for oriented sheets dried either freely or under restraint to 50% RH.

DISCUSSION

The hygroexpansion of paper is a result of the swelling of the fibrous material caused by the added volume of the sorbed water. The radial swelling of the fibre is several times greater than the longitudinal swelling, so the higher in-plane hygroexpansivity in the CD and the very high out-of-plane hygroexpansion are expected results (Figs. 5, 7 and 8). However, other structural factors, particularly effects due to drying restraints, also have a great influence on the hygroexpansivity.

Drying restrictions in the RH range, i.e. between 20% and 0% moisture content have a profound influence on the hygroexpansivity of paper (Figs. 3,4). This behaviour is contrary to that of the elastic modulus, which is mainly influenced by drying restrictions in the range of 40-70% solids content (4). In the case of the elastic modulus, the effects are related to the "freezing" of disordered zones in the microfibrils (4) whereas the effect on hygroexpansivity may be tied to a softening of the amorphous matrix material, mainly the hemicelluloses (5). In practice this means that the drying history has a greater influence on the hygroexpansivity of a paper sheet than on its mechanical properties.

To explain the influence of density of the in-plane hygroexpansivity the following is proposed: The number of fibre to fibre interfaces increases with increasing density. For freely dried sheets, this means that the transverse shrinkage of the crossing fibres is more effectively transmitted to shrink the sheet, which in turn leads to a higher in-plane hygroexpansivity (1) (Fig. 7). Naturally, for oriented sheets this effect becomes more prominent in the CD and almost negligible in the MD. For sheets dried under restraint, where shrinkage is prevented, increased density i.e. increased adhesion and increased drying forces leads to a higher orientation of the disordered regions of the microfibrils. This will in turn lead to a higher elastic modulus and produce swelling restrictions in the plane of the sheet, resulting in reduced in-plane hygroexpansivity.

Another important question to answer is how the volume hygroexpansion of paper relates to drying restraints and sheet density. The porous structure of paper must then be taken into account (6). For isotropic structural materials, voids such as pores, expand and contract to the same extent as the surrounding material. In a porous structure with expansional restrictions, swelling may be directed inwards to decrease the expansion of the voids. This is generally the case for anisotropic materials, where expansional restrictions often exist in certain directions. The wood fibre, with its wound structure of microfibrils at different angles, is a prime example of such a material. For example, it is well known that wood fibres with reasonably intact outer fibre wall layers swell inwards towards the lumen in high swelling conditions.

In a porous structure with expansional restrictions, such as paper, increased density should be expected to result in an increased volume expansivity. Such behaviour has been found for the thermal expansivity of polymeric foams (7). In the case of wood (8,9) and fibre building board (2), it has also been noticed that the volume hygroexpansivity is greater for more dense structures.

The void volume of wood is approximately constant during moisture sorption (9). For such a system the swelling of the material is related to the volume of the sorbed solvent. With increasing density the amount of sorptive material per unit volume increases and the volume expansivity, β'_V based on weight fraction (%/% weight increase in solvent fraction) will increase according to (9):

$$\beta'_V = \frac{\text{material (including pores)}}{\rho \text{ solvent}} \qquad (1)$$

For cellulosic materials, the maximum hygroexpansivity based on the volume of the sorbed water is thus about 1.5 %/% moisture ratio.

Tables 1 and 2 summarize the volume hygroexpansivity results which have been obtained in this study. Apparently, drying restraints do not influence volume hygroexpansivity at the higher densities. However, at the lower densities there is a very obvious effect (Table 2) in that the volume expansivity is greater for sheets dried under restraint and less for the freely dried sheets. The increase in the volume hygroexpansivity of the freely dried sheets with increasing density is very similar to the behaviour noticed for wood. Comparing the results with the calculated hygroexpansivity values from Equation 1, it is clear that (with one exception) the swelling has resulted in a slight increase in pore volume. It is also apparent that the magnitude of this increase is largely unaffected by the density of the sheet.

drying strategy according to Fig. 2	density ρ (kg/m^3)	volume expansivity β_v (%/% MC)	
		from β_{ip} and β_{op} according to equation (2)	from mercury method (12)
1. free	785	0.98	0.76
2. restr to 90 % RH	790	1.01	0.99
3. restr to 50 % RH	780	1.02	0.97
4. restr to 25 % RH	815	1.27	1.31

Table 1 Volume hygroexpansivity for isotropic sheets dried with varying degree of drying restraints ($23^{o}SR$, 100 g/m^2).

drying strategy according to Fig. 2	density ρ (kg/m^3)	volume expansivity β_v (%/% MC)	
		from β_{ip} and β_{op} according to equation (2)	calculated according to equation (1)
1. (freely dried)	327	0.24	0.33
	454	0.66	0.45
	560	0.68	0.56
	782	0.91	0.78
	909	1.16	0.91
3. (dried restrained to 50 % RH)	326	-	0.33
	447	1.37	0.45
	585	1.33	0.59
	800	1.13	0.80
	943	1.18	0.94

Table 2 Volume hygroexpansivity for oriented sheets wet-pressed to different densities and dried either freely or under restraint to 50 % RH ($17^{o}SR$, 250 g/m^2).

For the sheets dried under restraint, the greater expansivity at lower densities might be explained by irreversible effects, i.e. the release of stresses may permanently increase the thickness of the sheet. Nevertheless, the volume hygroexpansivity is always lower than the maximum hygroexpansivity (without expansional restrictions) of 1.5 %/% moisture ratio.

CONCLUDING REMARKS

The effects of density and drying strategy on the hygroexpansivity clearly exhibit some trends that are important to consider.

1. Drying restraints, particularly at high solids contents, have a great influence on the expansivity of paper.

2. For freely dried sheets an increase in density increases the in-plane hygroexpansivity in the CD as well as the out-of-plane hygroexpansivity.

3. For sheets dried under restraint, the density has only a marginal effect on the in-plane hygroexpansivity. The out-of-plane expansion probably involves irreversible structural changes, particularly at low densities.

EXPERIMENTAL DETAILS

The measurements were performed on laboratory sheets made from a commercial never-dried bleached pine sulphate pulp (Pinus silverstris). Isotropic sheets were made on a Finnish sheet mold at 100 g/m^2. Oriented sheets were made on a dynamic sheet former (Formette Dynamique CTP, France) with a grammage of 250 g/m^2. The oriented sheets were made under identical conditions which produced the same degree of fiber orientation. In the case of the restrained dried sheets, the ratio of the tensile strength in the MD to that in the CD was approximately 2.5. Sheets were wet-pressed to given densities and dried in drying frames (10) to the specified RH at 23°C.

The in-plane hygroexpansion was measured in a specially designed apparatus, in which the distance between two reference points about 120 mm apart on the paper strips was measured by a linear variable differential transformer (LVDT) to an accuracy of $\pm 1 \mu m$. The essential features of the apparatus are that the sample is placed horizontally and forced to be flat during the measurements, thus eliminating effects of curl and the need of forces acting in the plane of the sample.

The out-of-plane hygroexpansion is measured by evaluating the mean thickness of the sheets at different moisture contents. This is accomplished by sensing the thickness between two spherical platens with a diameter of 4.5mm under a load of 10g. The hygroexpansivity β is defined as the percentage change in dimension per percentage increase in moisture content by weight. For oriented sheets the in-plane hygroexpansivity is given by the geometric mean value, which has been shown to be a good measure of the isotropic in-plane hygroexpansivity (11).

The volume expansivity β_v is calculated from the out-of-plane (β_{op}) and the in-plane (β_{ip}) hygroexpansivities according to:

$$1 + \beta_v = (1 + \beta_{ip})^2(1 + \beta_{op}) \tag{2}$$

A mercury buoyancy technique which was originally developed by Wasser (12) was also used to determine the volume expansion.

ACKNOWLEDGEMENT

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Transcription of Discussion

The In-Plane and Out-of-Plane Hygroexpansional Properties of Paper

by L. Salmén, C. Fellers and M. Htun

Page You talk here about shrinkage and moisture expansion but I don't see any mention of microcompressions. I wonder if you attribute any of these effects to microcompressions or are you attributing all of the shrinkage and moisture expansion to changes in length of the cellulose fibril as a result of changes in the disordered region.

Dr. L. Salmén What we are looking at are the effects on the molecular level. When drying the paper freely so that microcompressions develop in the fibres, the molecules will of course be unrestrained and vice versa. The changes on the cellulose fibrils could be anything from changes in its angle to kinks or small length changes too small to appreciately affect the fibre length.

Marchessault How do you see that non-crystalline zone of the micro fibril acting? Your effect could be explained if you would agree that these zones are kinked and that the effect of tension is simply to straighten it out and then allow the neighbouring ones to cement and establish what I would call pseudo cross links. I don't quite understand what role you see this non crystalline domain playing. Incidentally, the hemicellulose would then be acting as a sort of glue.

Salmén The cellulose fibrils with its disordered zones we think are contributing both to strength properties and also to some extent to hygroexpansivity. They dominate the properties in the length direction of the fibrils and by stretching them you will get a stiffer fibre. This straining if it is a straightening or a pure elongation will anyhow put strain on the disordered zones thus reducing its mobility. The hygroexpansivity is mostly affected by the hemicellulose fraction and is acting most dominantly in the cross fibril direction. As I have shown, one has to consider two different mechanisms of property development when drying.

Back In the data presented on the out of plane hygroexpansivity you apply restraint on drying in plane. If, on the other hand, you apply Z direction restraint it will give you a higher out of plane hygroexpansivity. I think it would be worthwhile to carry out such an experiment for comparison.

Salmén Yes, I think that would be a good idea. The normal way of introducing Z-directional restraints of course also involves an in plane restriction so this result ties in with the results I have presented here.

Prof B. Steenberg Royal Inst. of Technology, Stockholm, Sweden

I believe there is considerable evidence that the hemicelluloses and similar degraded accessible material are the main components responsible for hygroexpansivity. Polymer resins filled with cellulose fibres, randomly oriented, show less hygroexpansivity the higher the alpha-cellulose content the fibres have. This is well known for urea formalde hyde as well as phenolics. Saturating papers for phenolic laminates made with extremely small additions of aliphatic long chain quarternary amines produced products with about one half the hygroexpansivity of paper made on the paper machine under the same conditions, but without the additive. The amines destroy paper strength.

Salmén I should perhaps point out that we are saying that the hygroexpansivity not only depends on the amount of hemicelluloses, but also on the degree of orientation or restraint of the hemicelluloses.

Ebeling May I recommend that those who have not already done so should read the 1961 publication by Page and Tydeman, in which they describe how microcompressions affect the dimensional stability of paper, only then should one consider if there is anything worthwhile to add to the subject.

Salmén What I have been talking about here is the effects on the molecular level. These effects do, of course, manifest themselves in effects on the macroscopic level.