JOURNAL OF MATERIALS SCIENCE 29 (1994) 2373–2377

General formula for hygroexpansion of paper

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A general formula for the hygroexpansion of paper has been derived. The hygroexpansion of paper is determined by two factors: one is the hygroexpansion of a single fibre, including the hygroexpansions in the fibre axis and the transverse directions; the other is the efficiency of the stress transfer from the network to the fibres when paper is subjected to uniaxial stress. The latter factor is dependent on mechanical properties of the fibre and the network. Because of its general nature and its simple physical significance, the formula can be used to interpret various experimental results in a qualitative or semi-quantitative manner. Some specialized cases are presented to illustrate unique hygroexpansion characteristics of bonded fibre networks. Effects of fibre orientation and the degree of fibre-to-fibre bonding are discussed based on experimental data and the derived formula. It is shown that the hygroexpansion of paper in the machine direction is almost entirely controlled by the hygroexpansion of fibre in the fibre-axis direction, whereas the hygroexpansion in the cross-machine direction is inherently sensitive to changes in the degree of fibre-to-fibre bonding and fibre orientation, because of the larger contribution of the hygroexpansion of fibre in the transverse direction.

1. Introduction

Paper consists of fibres highly oriented in the sheet plane direction and bonded together to form a porous random network (Fig. 1). Hygroexpansivity, defined as the dimensional changes due to the change in the relative humidity of the surrounding atmosphere, is generally a complex function of paper structure and the hygro-elastic properties of fibres.

The dimensional change of a single fibre is highly anisotropic. Although there are no hygroexpansion data of a single fibre available at this moment, it has been shown that shrinkage in the fibre axis direction during drying is of the order of 1–2%, whereas shrinkage in the transverse direction of a fibre is in the range of 20–50% [1]. When moisture content or relative humidity changes, the dimensional changes of a fibre are transmitted to neighbouring fibres through the bonded fibre network (Fig. 1). The degree of transfer of the dimensional changes from one fibre to another may be affected by a number of factors, such as the degree of fibre-to-fibre bonding, curliness of the fibres, fibre dimensions, orientation of the fibres, and stiffness ratio between the fibre axis and the transverse directions. Because of the complex structure of the network and interactions between mechanical properties and hygroexpansion, it is generally difficult to interpret experimental results of hygroexpansion in terms of paper structure and fibre properties.

In this paper, a general formula is developed relating fibre hygroexpansion to paper hygroexpansion. The general expression for multiphase elastic media obtained by Dvorak & Benveniste [2] can be easily transformed to a simple, physically interpretable form in the case of the bonded fibre network. Unique hygroexpansion characteristics of bonded fibre networks are illustrated by specializing the derived formula. Experiments are performed to determine effects of fibre orientation and the degree of fibre-to-fibre bonding. The relative contribution of hygroexpansion of fibre in the axial and in the transverse directions is discussed, using the experimental results and the derived formula.

2. General expression for hygroexpansion of bonded-fibre network

Dvorak & Benveniste [2] recently obtained a general equation relating local and overall 'eigen strain', such as thermal and hygro strains, for composite materials consisting of many distinct elastic phases:

$$\mu_{ij} = \frac{1}{V}\sum_p \int_{v_p} [B^p_{ijkl}(x)]^T \mu^p_{kl}(x)\mathrm{d}V \qquad (1)$$

where μ_{ij} and μ^p_{ij} are the overall and local eigen strains, which in our context are hygroexpansion strains of paper and fibre, respectively. V is the total volume of the representative volume element, the superscript T denotes the transpose of tensor, and B^p_{ijkl} is the mechanical influence function for the pth phase, defined as

$$\sigma^p_{ij}(x) = B^p_{ijkl}(x)\,\sigma_{kl} \qquad (2)$$

This is the tensor relating the overall stress σ_{kl} to the local stress $\sigma^p_{ij}(x)$ of the pth phase when the composite is subjected to the traction force at the surface which creates the overall stress σ_{kl}. In the case of the bonded fibre network, the stress in the pore volume is apparently zero, and therefore the corresponding influence function for the pore phase vanishes.

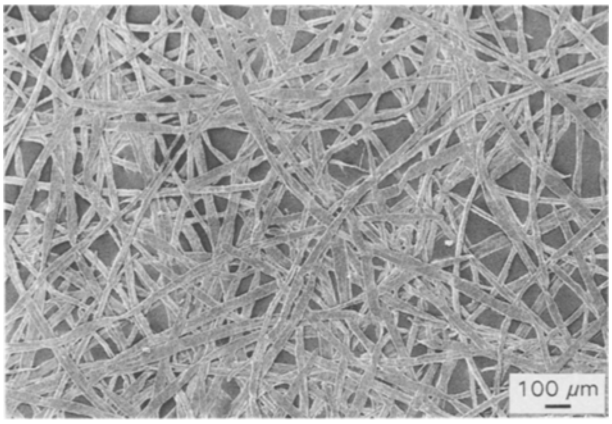

Figure 1 Paper as bonded-fibre network. Basis weight, 5g m^{-2}.

As the above equations are expressed in global coordinates, it is not easy to see the physical significance of each parameter when the anisotropic constituents have three-dimensional orientations, as typically seen in paper and non-woven fabric. To express the equation in the local coordinates embedded in the fibre element, as shown in Fig. 2 in the two-dimensional case, we first define the directional cosine l_{ij}, which transforms the global coordinate system into the local coordinate system. The stress components observed in the local coordinates are

$$\begin{aligned} \sigma^f_{ij}(x) &= l_{ir}(x)\, l_{js}(x)\, \sigma^p_{rs}(x) \\ &= B^p_{rskl}(x)\, \sigma_{kl}\, l_{ir}(x)\, l_{js}(x) \qquad (3) \\ \langle \sigma^f_{ij}(x) \rangle_V &= F_{ijkl}\sigma_{kl} \end{aligned}$$

where

$$F_{ijkl} = \langle B^p_{rskl}(x)\, l_{ir}(x)\, l_{js}(x) \rangle_V \qquad (4)$$

As seen in the above definition, F_{ijkl} is a tensor which relates the macroscopic stress to the average fibre stress observed in the principal directions of fibres. (The principal directions of a fibre are fibre axis, fibre width and fibre thickness directions.) For the case where moisture change (or temperature change) is uniform for all fibres, we can rewrite Equation 1 as

$$\begin{aligned} \mu_{ij} &= \langle [B^p_{ijrs}(x)]^T [l_{rk}(x)]^T [l_{sl}(x)]^T \mu^f_{kl} \rangle_V \\ &= (F_{ijkl})^T \mu^f_{kl} \qquad (5) \end{aligned}$$

where μ^f_{kl} is the hygroexpansion measured in the local coordinates taken along the principal directions of fibre (Fig. 2). Assuming orthotropic hygro-elastic properties of both fibre and paper, and taking the global coordinate system along the principal directions of paper that is, the machine, the cross-machine, and the paper thickness directions, we obtain

$$\begin{aligned} \mu_{11} &= F_{1111}\, \mu^f_{11} + F_{2211}\, \mu^f_{22} + F_{3311}\, \mu^f_{33} \\ \mu_{22} &= F_{1122}\, \mu^f_{11} + F_{2222}\, \mu^f_{22} + F_{3322}\, \mu^f_{33} \qquad (6) \\ \mu_{33} &= F_{1133}\, \mu^f_{11} + F_{2233}\, \mu^f_{22} + F_{3333}\, \mu^f_{33} \end{aligned}$$

It should be noted that in the above derivation there is no assumption made on fibre shapes, fibre dimensions and fibre orientation. Therefore, Equation 6 is valid for a general bonded fibre network consisting of curly, three dimensionally oriented fibres. It should also be noted that although the above equation was derived for the bonded fibre network whose constituent element is 'fibre', the same relation holds when the constituent elements are microfibrils, as long as the constituent materials retain the 'orthotropic' hygro-elastic properties.

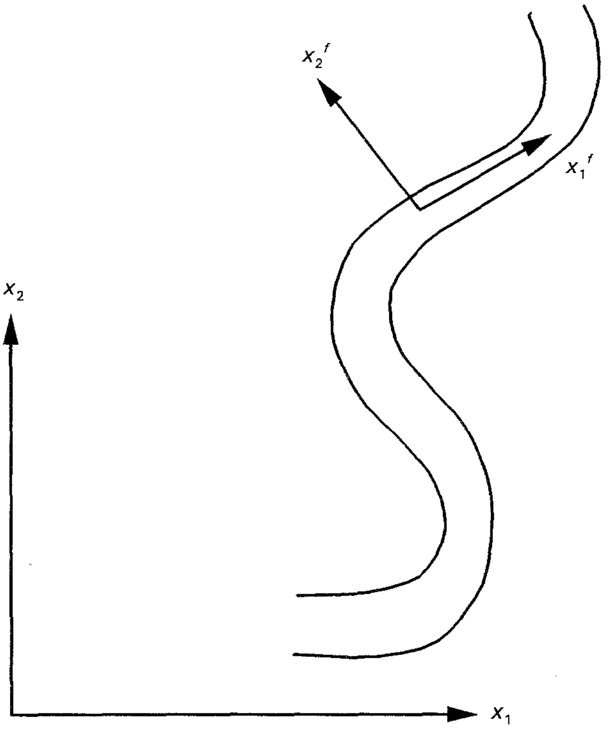

Figure 2 Local coordinates taken along the principal directions of fibre (two-dimensional case). The principal directions are generally functions of the position along the fibre length because of the fibre curl.

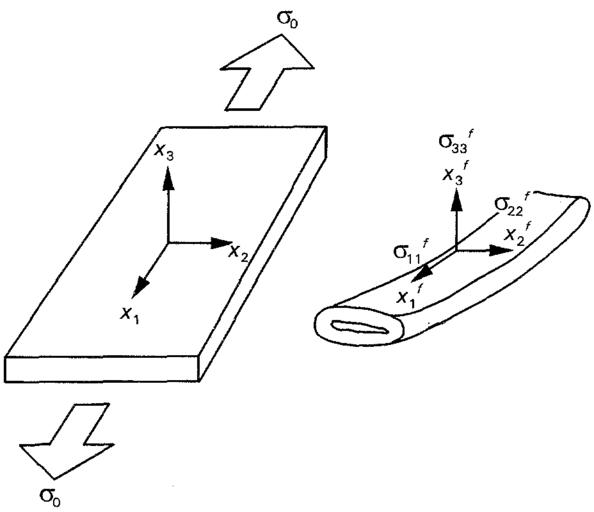

Figure 3 Uniaxial stress is transferred through the network to a fibre in the three principal directions: fibre axis, fibre width and fibre thickness directions.

The physical significance of F_{ijkl} can be easily found from the last part of Equation 3. When the uniaxial stress σ_0 is applied in the x_1 direction of paper (Fig. 3), the stress is transmitted to the fibres in the sheet in different degrees depending on fibre orientation, fibre bonding and geometrical shapes of each fibre. The

F_{1111} component is defined as

$$F_{1111} = \frac{\langle \sigma^f_{11} \rangle_V}{\sigma_0} \quad (7)$$

That is, F_{1111} is the ratio of the average stress transferred in the axial direction of the fibres $\langle \sigma^f_{11} \rangle$ to the applied stress σ_0. The F_{2211} and F_{3311} components can be defined in a similar way

$$F_{2211} = \frac{\langle \sigma^f_{22} \rangle_V}{\sigma_0}$$
$$F_{3311} = \frac{\langle \sigma^f_{33} \rangle_V}{\sigma_0} \quad (8)$$

The parameter F_{ijkl} therefore represents the efficiency of the stress transfer from the network to the three principal directions of the fibres. Hereinafter, we will call F_{ijkl} the stress transfer coefficient. From the invariant property of stress tensor:

$$\sigma_{11} + \sigma_{22} + \sigma_{33} = \langle \sigma^f_{11}(x) \rangle_V + \langle \sigma^f_{22}(x) \rangle_V + \langle \sigma^f_{33}(x) \rangle_V \quad (9)$$

F_{ijkl} satisfies the following equations:

$$F_{1111} + F_{2211} + F_{3311} = 1$$
$$F_{1122} + F_{2222} + F_{3322} = 1 \quad (10)$$
$$F_{1133} + F_{2233} + F_{3333} = 1$$

In the case of a random fibre network, calculation of the stress transfer coefficient is not easy either by direct elasticity analysis or by numerical methods. However, because of its simple physical significance, the stress transfer coefficient can be used for general consideration of the hygroexpansion responses of various networks. It should be emphasized that the stress transfer coefficient depends only on mechanical properties of the network, so that all the effects of the network geometry and fibre mechanical properties on hygroexpansivity of paper can be discussed based on this parameter without considering the complex interaction with hygroexpansion of fibres.

Using Equations 6, 9 and 10, we obtain another form of the formula, for example for a component μ_{11}:

$$\mu_{11} = \mu^f_{11} + (\mu^f_{22} - \mu^f_{11})\frac{\gamma_2}{1 + \gamma_2 + \gamma_3} + (\mu^f_{33} - \mu^f_{11})\frac{\gamma_3}{1 + \gamma_2 + \gamma_3} \quad (11)$$

where γ_2 and γ_3 are defined as

$$\gamma_2 = \frac{\langle \sigma^f_{22} \rangle_V}{\langle \sigma^f_{11} \rangle_V}, \quad \gamma_3 = \frac{\langle \sigma^f_{33} \rangle_V}{\langle \sigma^f_{11} \rangle_V} \quad (12)$$

The above equations clearly indicate that the hygroexpansion of paper is affected by the ratio of the average stress between the axial and the transverse directions of fibre.

3. Discussion

Unique characteristics of the hygroexpansion response of the fibre network can be revealed by specializing the general formula in Equation 6. The first two cases considered below were predicted previously by using the two dimensional model [3], but we will show that the same results can also be obtained in the general case of three dimensions.

3.1. Isotropic fibre

If the fibre hygroexpansion is isotropic:

$$\mu^f_{11} = \mu^f_{22} = \mu^f_{33} = \mu_0 \quad (13)$$

it can be shown by using Equations 6 and 10 that

$$\mu_{11} = \mu_{22} = \mu_{33} = \mu_0 \quad (14)$$

Thus, for the fibre network consisting of fibres with the isotropic hygroexpansion property, the overall hygroexpansion is isotropic and is exactly equal to fibre hygroexpansion. There is no effect of density (or pore volume), fibre dimensions and fibre mechanical properties on the overall hygroexpansivity.

3.2. Pin-pointed network

One of the unique features of the fibre network is that because of its structure and fibre dimensions, the stress is transferred preferentially to the fibre axis direction rather than to the transverse direction. In the extreme case when fibres are pinned (bonded) together at only one point in each bond site, such as the case of the pin-pointed network considered by Page and Tydeman [1], the applied stress is transferred only in the fibre axis direction

$$\langle \sigma^f_{22} \rangle_V = \langle \sigma^f_{33} \rangle_V = 0 \quad (15)$$

Using the last part of Equation 3, we can show that

$$F_{22ij} = F_{33ij} = 0 \quad (16)$$

Therefore, Equations 16 and 6 give

$$\mu_{11} = \mu_{22} = \mu_{33} = \mu^f_{11} \quad (17)$$

Again, the hygroexpansivity of the pin-pointed network is not affected by the network structure, fibre dimensions, fibre mechanical properties and degree of fibre bonding, but is fully determined by the hygroexpansion of fibre in the fibre-axis direction.

3.3. Transversely isotropic fibre

A more realistic example may be obtained by assuming that the fibre is transversely isotropic in its hygroelastic properties, that is, the properties in the fibre axis direction are different from those in the transverse plane direction, but within the transverse plane the properties are isotropic. In other words,

$$\mu^f_{11} = \mu^f_L, \quad \mu^f_{22} = \mu^f_{33} = \mu^f_T \quad (18)$$

Then Equation 6 can be rewritten for the in-plane hygroexpansion by using Equation 10 as

$$\mu_{11} = \mu^f_L + b_1(\mu^f_T - \mu^f_L)$$
$$\mu_{22} = \mu^f_L + b_2(\mu^f_T - \mu^f_L) \quad (19)$$

where

$$
\begin{aligned}
b_1 &= 1 - F_{1111} = F_{2211} + F_{3311} \\
b_2 &= 1 - F_{1122} = F_{2222} + F_{3322}
\end{aligned} \tag{20}
$$

As seen in the above definition, the parameter b_1 represents the degree of stress transfer to the transverse directions of fibre when the sheet is tensioned in the x_1 direction, and the parameter b_2 is defined in the same way when the sheet is tensioned in the x_2 direction. Both parameters are generally functions of fibre orientation, fibre stiffness, the degree of fibre-to-fibre bonding, and fibre dimensions. Under the assumption that the constituent fibres are randomly oriented in the plane direction and applied strains are uniformly transmitted to the fibres, it was shown that b_1 (or b_2) is equal to the ratio of the transverse stiffness to the axial stiffness of a fibre [3].

The simplest example is the effect of fibre orientation. Let us take the x_1 and x_2 directions as the machine direction and the cross-machine direction of paper, respectively. As more fibres orient in the machine direction, the stress is transferred more in the axial direction of the fibres when the sheet is subjected to tension in the machine direction (F_{1111} increases). This results in a decrease in the parameter b_1 (the first part of Equation 20), and thus causes a decrease in hygroexpansion in the machine direction (μ_{11}) according to Equation 19. The parameter b_2, on the other hand, increases with increasing fibre orientation according to its definition (the second equation of Equation 20), that is, with increasing fibre orientation in the machine direction, the stress transfer in the fibre axis direction decreases when the tension is applied in the cross-machine direction (F_{1122} decreases), and thus b_2 increases with increasing fibre orientation in the machine direction. Therefore, the hygroexpansion of the sheet in the cross-machine direction (μ_{22}) increases according to Equation 19.

Figs 4 and 5 show an example of the effect of fibre orientation on the hygroexpansion anisotropy. Hygroexpansion coefficients were determined from the slope of the linear part of hygro-strain against moisture content curves according to the procedure described in [4]. Sheets with different fibre orientations were made using a dynamic sheet former from an unbleached, unbeaten, softwood kraft pulp. The sheets were dried with and without restraint. For freely-dried sheets (Fig. 4), with increasing elastic stiffness ratio (or with increasing degree of fibre orientation), the hygroexpansivity in the machine direction decreases whereas the hygroexpansivity in the cross machine direction increases, as expected from the above discussion. It was also found that the effect of density on the relationship between hygroexpansivity and the stiffness ratio is much pronounced in the cross-machine direction as compared with that in the machine direction, that is, the cross-machine direction hygroexpansivity is very sensitive to changes in the degree of fibre-to-fibre bonding. Fig. 5 shows the results for sheets dried under restraint. The general trend was the same as that for freely-dried sheets. It is interesting to note that the machine-direction hygroexpansivity, in particular for the low density sheets, does not show significant dependence on the degree of fibre orientation.

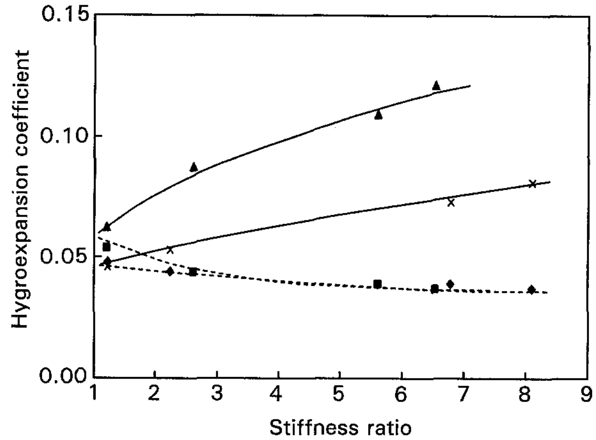

Figure 4 Effect of fibre orientation on hygroexpansivity for freely-dried sheets made from a fines-free, unbeaten, unbleached softwood kraft pulp (UBKP). Degree of fibre orientation expressed as the elastic stiffness ratio determined by an ultrasonic propagation method. Density levels were varied by changing wet pressing pressure. High density, 419 kg m^{-3}; low density, 220 kg m^{-3}. Cross-machine direction, ▲, high; ×, low density. Machine direction, ■, high; ◆, low density.

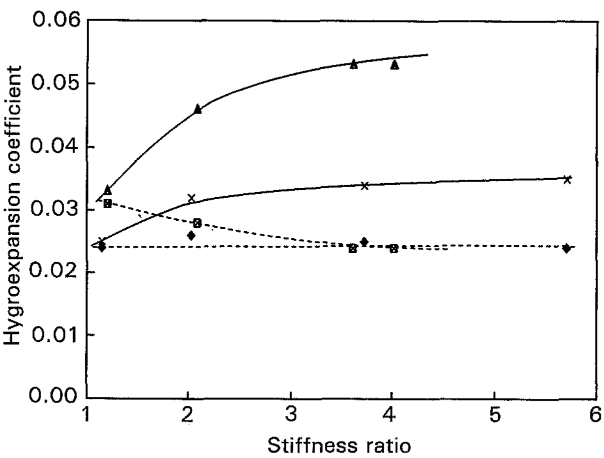

Figure 5 Effect of fibre orientation on hygroexpansivity for restraint-dried sheets made from a fines-free, unbeaten, unbleached softwood kraft pulp (UBKP). Degree of fibre orientation expressed as the elastic stiffness ratio determined by an ultrasonic propagation method. Density levels were varied by changing wet pressing pressure. High density, 419 kg m^{-3}; low density, 220 kg m^{-3}. Cross-machine direction, ▲, high; ×, low density. Machine direction, ■, high; ◆, low density

The results presented in Figs 4 and 5 suggest an important mechanism controlling the hygroexpansion of paper. In view of Equation 19, the hygroexpansion of paper consists of two contributions: one is from the hygroexpansion in the fibre axis direction (the first term), and the other is from the hygroexpansion in the transverse direction (the second term). The effect of the latter term depends on the paper structure, including fibre orientation and the degree of fibre-to-fibre bonding, through the parameters b_1 and b_2. Very little dependency of the machine-direction hygroexpansivity on the fibre orientation and the degree of bonding, as seen in Figs 4 and 5, suggests that the machine-direction hygroexpansivity is almost entirely controlled by the first term, i.e. the hygroexpansivity in the fibre-axis direction. The cross-machine direction hygroexpansivity is, on the other hand, more sensitive to changes in fibre orientation and the degree of fibre-to-fibre bonding because of the larger contribution of the second term in Equation 19. As the hygroexpansion in the transverse direction of a single fibre far exceeds the hygroexpansion in the fibre axis direction [1], it is often said that the hygroexpansivity of paper is determined by the transverse hygroexpansivity of fibre. However, Figs 4 and 5 show that the contribution of the hygroexpansion in the fibre axis direction is not small, amounting to 30–40% even in the case of the cross-machine hygroexpansion for the highly oriented sheets. This is expected by considering the unique stress-transfer characteristics of the bonded-fibre network, as discussed previously in the case of the pin-pointed network. Because of the elastic anisotropy of pulp fibre and the slenderness of its shape, the parameters b_1 and b_2 take small values, significantly reducing the effect of the transverse hygroexpansion of fibre (the second term of Equation 19).

For the out-of-plane hygroexpansion, Equation 6 gives

$$\mu_{33} = \mu_T^f - F_{1133}(\mu_T^f - \mu_L^f) \qquad (21)$$

As paper shows a highly oriented and layered structure in the plane of the sheet, F_{1133} may generally be small. As the fibres orient more in the plane direction, F_{1133} apparently decreases, and thus the hygroexpansion in the thickness direction of paper is expected to increase according to Equation 21, while the in-plane hygroexpansion decreases (see also Equations 19 and 20). This seems to be the case when drying restraint is applied to the sheet. In fact, Baum *et al.* [5] found that straining wet sheets increases the in-plane orientation of fibres. Salmén *et al.* [6] showed that the out-of-plane hydroexpansivity is higher for restraint-dried sheets than for freely-dried sheets.

4. Conclusions

The complex network geometry and interactions between mechanical and hygroexpansion properties of fibres often hinder physical understanding of the hygroexpansion mechanism of paper. The formula presented allows consideration of these mechanical and hygroexpansion effects separately. It has a very simple physical significance, and can be applied to a general network consisting of fibres with various shapes and three dimensional orientations.

Acknowledgement

The author thanks Ms C. Moss for her skilful experiments on the effect of fibre orientation on hygroexpansivity.

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Received 16 April
and accepted 19 October 1993