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INFLUENCE OF TEMPERATURE AND HUMIDITY ON THE ELASTIC AND EXPANSIONAL PROPERTIES OF PAPER AND THE CONSTITUENT FIBRE

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Synopsis The influence of humidity and temperature on the change in elastic properties of single fibres and the corresponding papers are shown to be similar.

Furthermore, expansional changes due to variation in the atmospheric conditions are shown to be related to the elastic properties of the sheet. For thermal expansion in paper, the product of coefficient of linear expansion and the elastic properties is approximately constant in all directions. A similar relationship is also followed in hygroexpansion. In the experiments reported, a slightly higher value in the machine-direction was obtained for the product of the dimensional changes and the elastic modulus.

It is also noted that the geometric means of the dimensional instability and elastic properties in machine and cross directions coincide with the same properties of the isotropic sheet.

Hygrodimensional changes for single fibres were measured by means of following the rotational twist of the fibres caused by sorption. It is deemed from these experiments that the fibre swelling is proportional to the moisture regain.

Finally, it is shown that the viscoelastic properties of cellulose material change in an irregular manner during changes of the moisture regain. These effects cause considerable reduction in the creep to failure time (lifetime) of paper in compression or tension under transient atmospheric conditions.

Introduction

THIS paper is intended to summarise experimental studies of the influence of humidity and temperature on the mechanical properties and dimensional instability of cellulose fibres and paper sheets. Clarification of the relationships between the elastic properties and dimensional changes in the materials investigated are also attempted.

Under the chairmanship of Dr H. G. Higgins

The objective with this work has been to demonstrate that the change of elastic properties of the sheet with changes in atmospheric conditions is essentially governed by the properties of the fibres. Moreover, it is intended to demonstrate that the influence of temperature and humidity are analogous in their effect on the elastic properties of cellulose materials. In general, the experimental results conform with these intentions.

Furthermore, it is shown that an interdependence exists between the elastic properties and the expansional properties of the sheet. Thus, a reorientation of the fibres in the sheet leading to a higher modulus in one direction is accompanied by a simultaneous decrease in hygroexpansivity.

Previous contributions on this subject have demonstrated that an interdependency exists and from a pure fundamental analysis of specific systems a qualitative relationship can be derived.[1] In paper, however, the situation is complicated by its network character, making it difficult to quantify the relative importance of the properties of the individual fibres, of the network structure and of the state of bonding.

Problems of similar complexity are being approached in the field of composite materials. In many cases, both the theoretical and experimental developments in this area have proved useful in paper physics. Special mention should be given to the recent effort by Mark,[2] Page[3] and several other authors to calculate the elastic properties of fibres. Likewise, Barkas[4] and Warburton[5] have derived equations to calculate the elastic properties of wood from the changes in moisture regain during fluctuations in relative vapour pressure under specified hydrostatic or directional stresses. It is not possible to express the relationship between equilibrium expansion and elastic properties with Barkas' approach. Hence, Rosen[6] claimed that this demands the introduction of unspecified internal restraints.

Halpin[7] recently suggested theoretical means to express the dependence of expansion on the orientational and elastic properties of orthotropic plates. An essentially similar approach to the twisting of single fibres has been taken by Mark.[8] In the present paper, some of the equations of Halpin and Mark are used to test their relevancy to the experimental data.

Influence of temperature on rigidity and expansion of sheet and fibres

Sheet properties

THE influence of temperature in a dry nitrogen atmosphere on the coefficient of thermal expansion for a kraft paper is shown in Fig. 1. As shown, an increase in the coefficient of thermal expansion occurs over the investigated temperature interval. The changes in the coefficient of thermal expansion is stepwise, indicating the presence of secondary transitions.

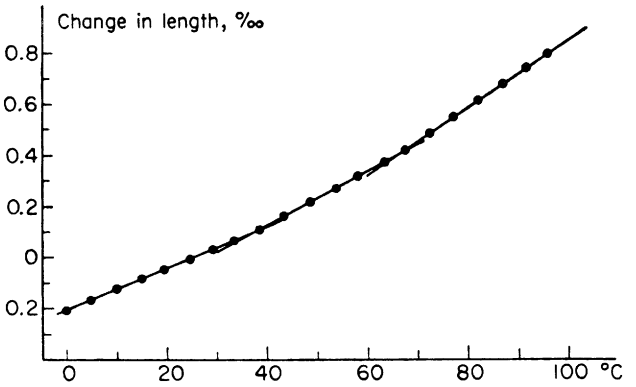

Fig. 1—The change in length with temperature for an unbleached sulphate paper (47 per cent)

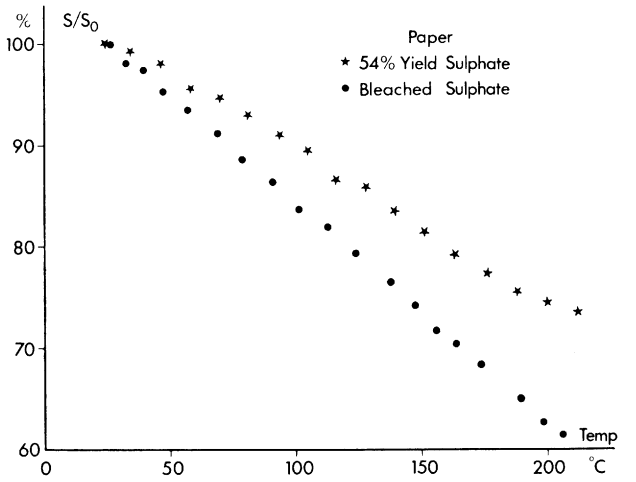

Fig. 2—The change in relative rigidity of paper with temperature

The presence of secondary transitions in cellulose and in the other wood polymers has previously been suggested by Goring and many other authors.[9–12]

The effect of temperature on the torsional rigidity of dry sheets of paper is illustrated in Fig. 2. Depending upon the type of raw material, a slightly different appearance of the curve is obtained. At higher temperatures, there is a tendency of the high lignin content pulps to have a higher rate of reduction of rigidity with temperature. The relative decrease of the rigidity with temperature is significantly higher for the bleached pulp.

This data was obtained in torsional experiments, but the temperature dependence is analogous to that reported by other authors for the elastic properties in tension.[14, 15] The relative change in elastic properties with temperature is also in agreement with the theoretical predictions of Nissan.[16] He calculated the relative change in elastic modulus to be about 2×10^{-3}/°C. This is of the same order as found in the present measurements.

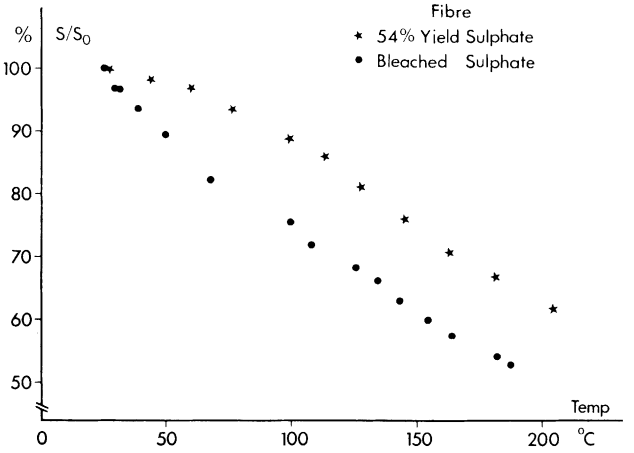

Fig. 3—The change in relative rigidity for single fibres with temperature

Single fibre properties

The same type of measurement has also been performed on single fibres obtained from the same pulps. In Fig. 3, the variation of torsional rigidity of single fibres is illustrated as a function of temperature in a nitrogen atmosphere. Absolute numbers for the shear modulus have been calculated at room temperature for a few kinds of fibre and the results are given in Table 1.

TABLE 1—MECHANICAL PROPERTIES OF SINGLE FIBRES

Sample	*Yield, per cent*	*Fibrillar angle X-ray*	*Fibrillar angle Polar†*	*Shear modulus, dyne/cm² × 10¹⁰*	*Breaking load, g*
Late wood—					
Unbleached sulphate	54	9°	1·7°	1·2	28·2±13
Unbleached sulphite	58	8°	6·0°	1·5	24·9±10
Ramie				2·0	
	—	8°	—	(1·7)*	23·2±12·4

* R. Meredith[37] † Polarised light measurements

The change in the torsional rigidity of single fibres with temperature is quite analogous to the results obtained for sheets. Young & Haugen[17]

measured the bending stiffness of fibres and obtained relatively larger changes in the elastic modulus, but it seems that his measurements overemphasise the effect of temperature.

Effect of fibre orientation on elastic properties and thermal expansivity of sheets

THE values of the coefficient of linear thermal expansion of various paper grades investigated are summarised in Table 2, together with the corresponding values of the modulus of elasticity. In all cases, the measurements refer to dry samples at 25° C. Expansion, like the modulus of elasticity, shows a pronounced directional dependence. The thermal expansivity reaches its highest value in the cross-direction, whereas the modulus is highest in the machine-direction.

The product $E\lambda$, given in Table 2, remains approximately constant for machine-direction and cross-direction samples, but the value of the product varies among grades.

Furthermore, measurements taken on each five degree increment between machine and cross directions show that the product $E\lambda$ remains constant for all directions between the machine and cross directions.[18]

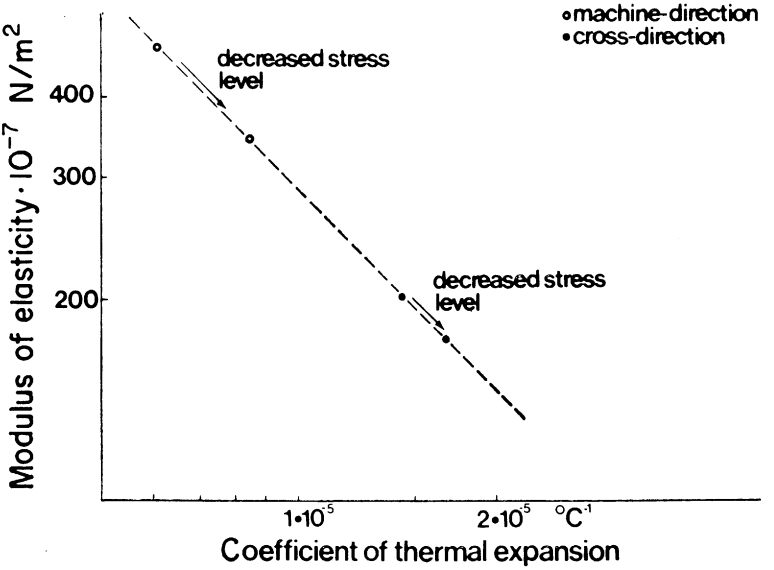

Fig. 4—The influence of destressing on the elastic modulus and coefficient of thermal expansion for an unbleached sulphate sheet (47 per cent) under dry conditions

The effect of releasing internal stresses is illustrated in Fig. 4, where log E has been plotted against log λ for kraft paper before and after a destressing treatment. Since the destressing process only moves the values for both machine and cross direction along the straight line of -1 slope in the double logarithmic diagram, the $E\lambda$ product remains constant.

TABLE 2—COEFFICIENT OF THERMAL EXPANSION AND MODULUS OF ELASTICITY OF VARIOUS PAPER GRADES

Paper grade	*Coefficient of thermal expansion,* (λ) $°C^{-1} \times 10^{-6}$		*E modulus,* kp/mm^2		*Product* $E\lambda \times 10^{-3}$	
	MD	*CD*	*MD*	*CD*	*MD*	*CD*
Greaseproof	7·5	15·5	795	416	5·96	6·45
MG kraft paper	6·4	13·4	720	267	4·61	3·58
Kraft sack paper	6·1	16·2	470	181	2·87	2·94
Newsprint	5·6	13·6	282	107	1·58	1·39
Fluting	7·4	12·1	276	196	2·04	2·37
White-lined duplex	5·9	12·3	263	105	1·55	1·29
Solid bleached board	4·2	8·7	347	152	1·46	1·34
White-lined chipboard	3·6	15·2	357	78	1·28	1·18
Bleached sulphate pulp	6·1	7·9	218	204	1·33	1·61

Effect of sheet density on thermal expansivity and elasticity

A SERIES of bleached sulphate pulps were prepared at different densities by wet pressing after beating. All sheets were destressed after drying by cycling between 35 per cent rh and 95 per cent rh for 7 days. The dependency of the modulus of elasticity on sheet density is illustrated in Fig. 5. Empirically, the dependency of the elastic modulus on density is given by the following equation—

$$E = k_1\rho^2 + k_2$$

where E = elastic modulus, N/m^2
ρ = sheet density, kg/m^3
k_1 = constant
k_2 = constant

This empirical result has previously been demonstrated by Luner.[19] It should be noted, however, that this 'square' dependency was found only for sheets destressed in the manner described.

In contrast with the behaviour of the elastic properties, the thermal expansivity does not show any marked dependency on density. No significant change in thermal expansivity could be found when the density was changed either by wet pressing or beating. The unbeaten pulp (15° SR) showed a small deviation in thermal expansivity from the beaten pulps. This might indicate that the properties of the fibres were changed by the mechanical treatment.

Apparently, the coefficient of thermal expansion is not dependent on sheet

density, suggesting that this property basically reflects the properties of the fibres in the sheet. Yet the elastic modulus depends strongly on sheet density. Consequently, it cannot be expected that the product $E\lambda$ is the same for various grades of paper with different densities.

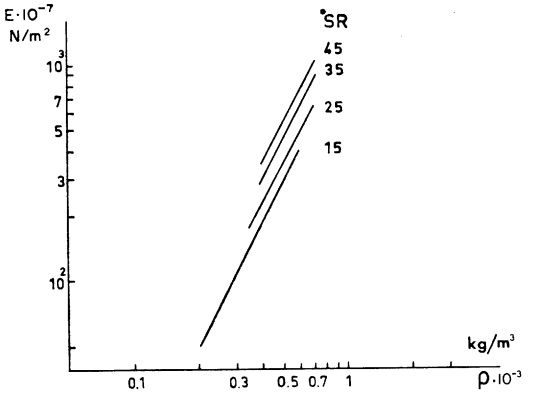

Fig. 5—Elastic modulus plotted against density (wet pressed) for bleached sulphate pulp at various levels of beating

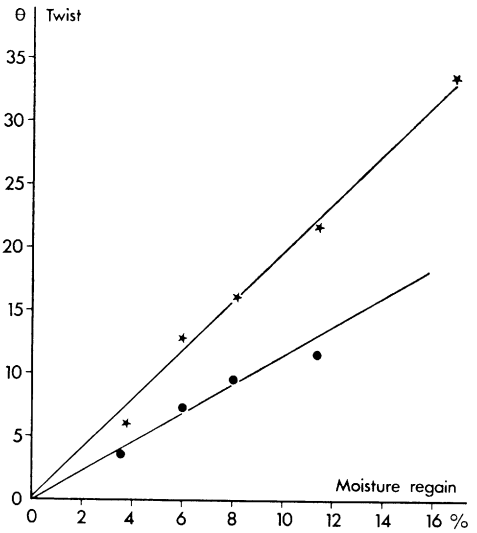

Fig. 6—The rotational twist θ (angular rotation) of single fibres (bleached sulphate) plotted against moisture regain

Dimensional changes due to humidity variations

THE dimensional changes in single fibres is experimentally difficult to follow, but previous investigations[20] have shown that the expansion in the longitudinal direction is small compared with that in the axial direction. In this investigation, it was found that the twisting displayed by the fibres was a convenient way to follow the dimensional changes in the fibres arising from changes in moisture regain.

In Fig. 6, the angular rotation of bleached sulphate fibres as a function of moisure regain is shown. According to the theoretical derivation made by Mark,[8] the twist angle should be proportional to the linear expansion of the fibres for various changes in the level of moisture content. The linearity between the rotational twist and the regain indicates that the linear expansion is constant at all humidity levels. This result agrees well with previous investigations, which have shown a proportionality between the fibre swelling in the radial direction and the moisture regain.[21]

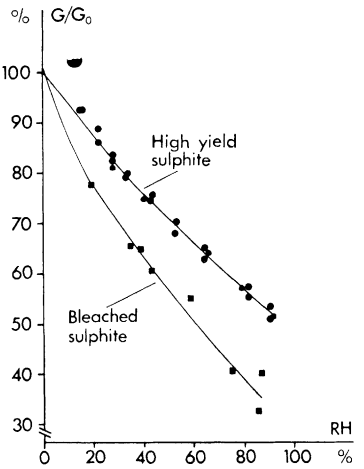

Fig. 7—The relative reduction in shear modulus for sulphite fibres with relative humidity: the result of several individual measurements is plotted

As noted, the fibres display a significant difference in twist, which should be attributed to the difference in microfibrillar angle. The formula of Mark allows for a correction because of the influence of the orientation of microfibrils. In this case, an individual determination of the microfibrillar angle was not made.

Considering the agreement with the theoretical derivation and the experimental results, it seems that the measure of twist is an elegant way to follow both the hygroexpansion of single fibres at constant fibril orientation and to measure fibrillar angle for a fibre population of the same raw material.

In several previous investigations, it has been documented that the hygroexpansion of paper is proportional to the moisture regain.[22]

Consequently, the expansional behaviour of the sheet is related to that of the fibres in the respect that the dimensional changes are proportional to the the moisture regain.

It is evident from the work of Brecht[23] and Nordman,[24] however, that processing variables such as drying conditions and beating have a large effect on the magnitude of the dimensional changes of the sheet.

Effect of humidity on the elastic properties of the sheet and single fibres

In Fig. 7, the variation in shear modulus with moisture regain for a high yield sulphite fibre and a bleached sulphite fibre is shown. As can be seen, the bleached fibre has a higher sensitivity towards changes in humidity than do the unbleached fibres.

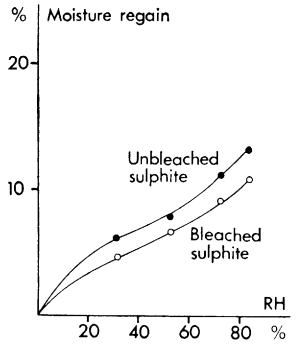

Fig. 8—Moisture regain with relative humidity for unbleached and bleached sulphite pulps

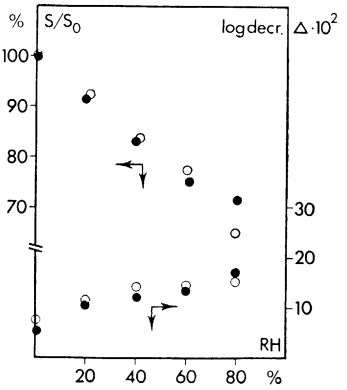

Fig. 9—The relative change in rigidity S/S_0 and the logarithmic decrement plotted against humidity for paper (●) and fibre (○) (sulphate 54 per cent yield)

This is analogous to the effect of temperature. Despite the larger change in shear modulus for the bleached fibres, the amount of moisture regain is slightly lower at the same humidity level. This can be seen in the absorption isotherms shown in Fig. 8. The general level of reduction in shear modulus is in agreement with the investigation of Haugen & Young,[17] but these authors measured the elastic modulus by means of bending experiments.

The relative changes in torsional rigidity and the logarithmic decrement for paper and corresponding single fibres are shown in Fig. 9. As seen, the effects of humidity on the mechanical properties of the sheet are fully analogous the effects on the single fibres. Considering the reduction in rigidity due to humidity changes for single fibres, it corresponds with the changes measured for sheets in this and other investigations.[25]

Thus, it seems reasonable to conclude that the network structure *per se* has little influence on the variations of the elastic properties of the sheet under different environmental conditions. Instead, the response to changes in atmospheric conditions is governed by the properties of the fibres.

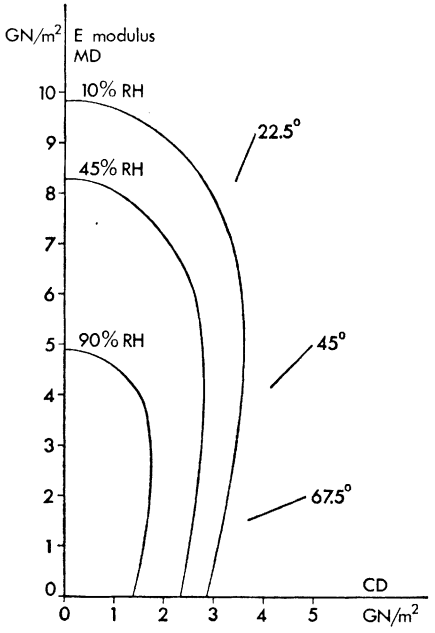

Fig. 10—Polar diagram of the elastic modulus for an oriented sheet (sulphate 47 per cent) at various humidities

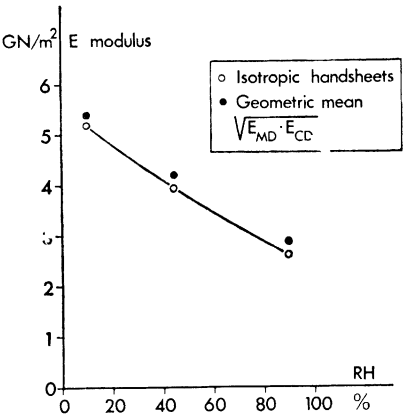

Fig. 11—Comparison of the elastic modulus for an isotropic sheet and the geometric mean ($\sqrt{MD \cdot CD}$) of an oriented sheet (sulphate 47 per cent)

Effect of humidity on the elastic properties and on the anistropy in hygroexpansion

THE influence of relative humidity on the elastic properties of a bleached sulphite sheet is illustrated by the polar diagram in Fig. 10, where it is also shown that the geometric means of the elastic modulus in the machine and cross directions both coincide with the elastic properties of the isotropic sheet.

Similar results have been obtained in previous investigations.[26] The relative humidity has a small effect on the elastic anisotropy in the sheet. In Fig. 12, the ratio of the elastic modulus in the machine and in other directions are given at various relative humidity levels. As can be seen, the anisotropy increases with relative humidity, indicating that the elastic modulus in the cross-direction is more sensitive to humidity changes than is the machine-direction modulus.

In Fig. 13, the hygroexpansion in different directions for sheets of an unbleached kraft pulp is shown. The hygroexpansion of an isotropic sheet of the same raw material is also shown on the same graph. These values coincide with the geometric mean of the hygroexpansion in the machine and cross directions of the oriented sheet. In comparison with the thermal expansion, the hygroexpansion is the highest in the direction in which the elastic property is the lowest. This result conforms with previous investigations by Brecht[23] and Prusas.[27]

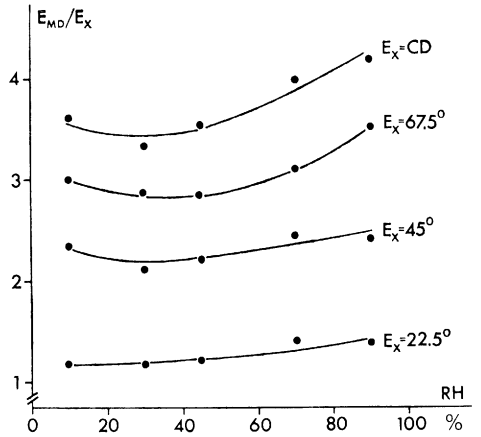

Fig. 12—The ratio of machine-direction modulus and elastic modulus in various directions plotted against relative humidity

The anisotropy of the hygroexpansion is slightly less pronounced than the anisotropy of the elastic properties. This is illustrated in Fig. 15, in which the elastic modulus is plotted against hygroexpansion. Apparently, the product of the hygroexpansion and elastic modulus is slightly higher in the machine-direction than in the cross-direction. Consequently, a direct analogy to the thermal behaviour (that is, $E \cdot \lambda = \text{constant}$) is not at hand for the hygroexpansion, but it has been established in this investigation and in previous experiments that the geometric means of the hygroexpansion in the MD and

CD are close to the hygroexpansion of the isotropic sheet, pressed and dried to the same density. Thus, it seems that the elastic and hygroexpansional potential in the sheet, regardless of the anisotropy, can be estimated from the evaluation of handsheets.

The expansion in various directions can be estimated also by the transformation equation given by Halpin[7] for the orientational dependence of expansional strains in an orthotropic plate.

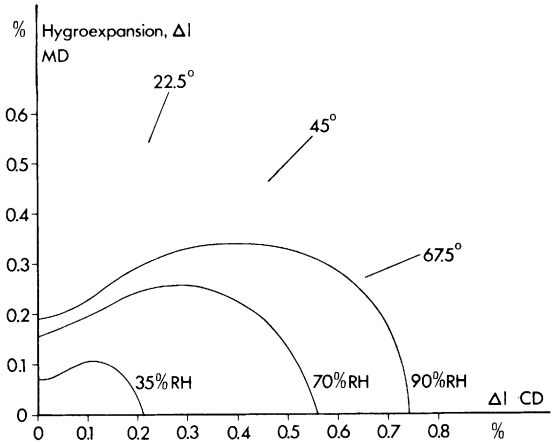

Fig. 13—Polar diagram of the hygroexpansion in an oriented and destressed sheet (sulphate 47 per cent) at various humidities

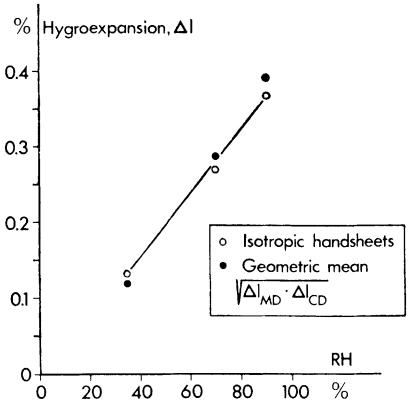

Fig. 14—Comparison of the hygroexpansion for an isotropic sheet and the geometric mean ($\sqrt{MD \cdot CD}$) of an oriented sheet

As can be seen from Table 3, good agreement is obtained, which indicates that the model used by Halpin is useful for estimating hygroexpansion in the various directions of oriented sheets.

TABLE 3—MEASURED AND CALCULATED HYGROEXPANSION FOR AN UNBLEACHED KRAFT

Relative humidity, per cent	*MD measured*	*Angle 22·5°*		*Angle 45°*		*Angle 67·5°*		*CD measured*
		Measured	*Calculated*	*Measured*	*Calculated*	*Measured*	*Calculated*	
10	0·00	0·00	0·00	0·00	0·00	0·00	0·00	0·00
35	0·07	0·08	0·09	0·14	0·14	0·19	0·19	0·21
70	0·16	0·20	0·22	0·36	0·36	0·49	0·50	0·56
90	1·19	0·23	0·27	0·48	0·47	0·68	0·66	0·74

Transient conditions

DURING transient environmental conditions, the logarithmic decrement of cellulose fibres changes in a manner that is described by the curve in Fig. 16. Meanwhile, a gradual decrease in shear modulus is experienced ranging from the equilibrium value measured at the lower relative humidity (rh) to that measured at the higher rh. As shown, however, the logarithmic decrement displays a maximum during the sorption of moisture, after which a gradual and slow decrease to the equilibrium value occurs. This indicates that during the sorption of the water a state of disequilibrium is attained for the viscoelastic properties of the cellulose material. Kubát[28] has previously reported this phenomenon for paper and other hygroscopic polymers. Other workers investigating wool fibres have documented similar behaviour during transient conditions, also reported disequilibrium behaviour of the elastic properties.[24]

The regain of moisture by single fibres is shown as a function of time in Fig. 17. The total water uptake is close to equilibrium after 1 h. Despite this fact, the logarithmic decrement of the fibres continues to change during the first 24 h and, according to Kubát, small changes continue to occur up to 85 h. This implies that the change in the logarithmic decrement represents the response of the solid system to the internal diffusion of water molecules. Evidently, the water molecules inside the hygroscopic gel are not initially placed at the energetically most favourable positions, but for a considerable time shift to positions that will yield a minimum of free energy.

Regardless of the underlying mechanism, the maximum in the logarithmic decrement illustrated in Fig. 15 indicates that the cellulose material will also have different properties with regard to other viscoelastic properties (for example, creep rate and stress relaxation) during internal diffusion of water molecules. Specifically, the rate of creep is important with regard to the lifetime of materials under a constant stress.

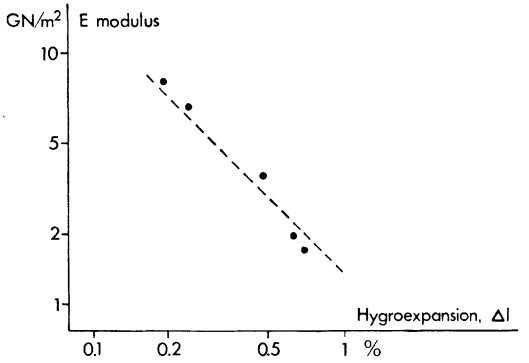

Fig. 15—Elastic modulus E at W per cent rh plotted against the hygroexpansion Δl between 10 and 95 per cent rh for a 47 per cent sulphate sheet

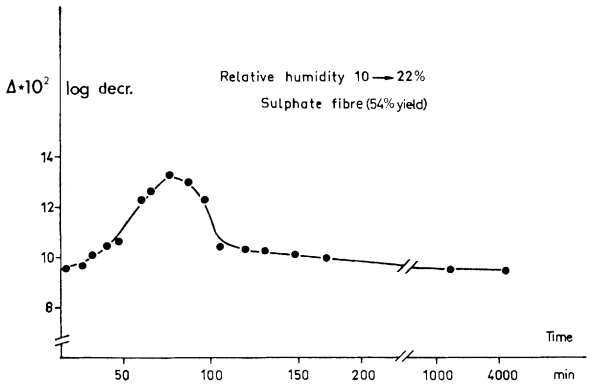

Fig. 16—Typical variation of the change of the logarithmic decrement with time during humidity change for cellulose fibres

The manner in which transient conditions influence the time to failure (or lifetime) in compression and in tension of paper or board are illustrated in Fig. 18 & 19. It can be seen that the time to failure is reduced when transient conditions prevail by comparing with the corresponding equilibrium conditions. This indicates that the rate of creep has increased considerably during the transient conditions.

The lowering of the time to failure during transient humidity conditioning has been reported by Byrd[30] and Lundberg & de Ruvo.[31]

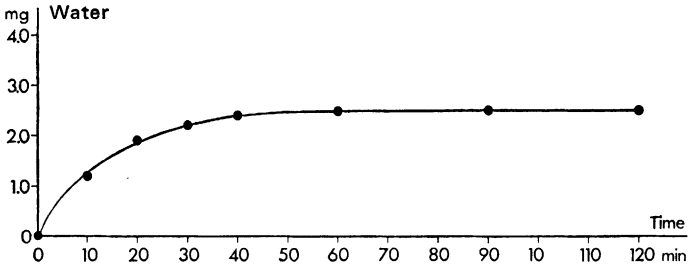

Fig. 17—Typical curve for the time dependency of moisture sorption for cellulose materials (sulphate fibre 54 per cent), 0→30 per cent rh

The first author, however, did not attribute these phenomena to intrafibre phenomena, but merely to changes in the interfibre bonding by the diffusion of water.

Conclusions

FROM the experimental results presented, the following concluding remarks can be made—

1. Temperature and humidity affect the elastic properties of the single fibres and the sheet in an analogous manner.
2. The elastic properties of bleached fibres have a higher sensitivity to temperature or humidity than do unbleached fibres, despite the larger amount of amorphous material in the latter variety of fibres.
3. Both fibres and paper expand in proportion to the amount of sorbed water.
4. Expansional strains and elastic properties are inversely related, but the anisotropy in hygroexpansion is not found to be as pronounced as the anisotropy in elastic properties.
5. Transient climatic conditions change the viscoelastic properties of both the fibres and the sheet. This is shown to lead to a reduced lifetime of paper during creep experiments.

These results indicate that a retention of elastic properties or rigidity during variations in the environment is best obtained when pulps of higher yield are used. It is evident that little in dimensional stability can be gained by reorientation in the sheet with regard to the general stability. Generally, drying stresses have to be introduced in the sheet to improve this property.

EXPERIMENTAL

Thermal expansivity

THE coefficient of thermal expansion of paper was determined using a technique described by Martin-Löf *et al.*[18]

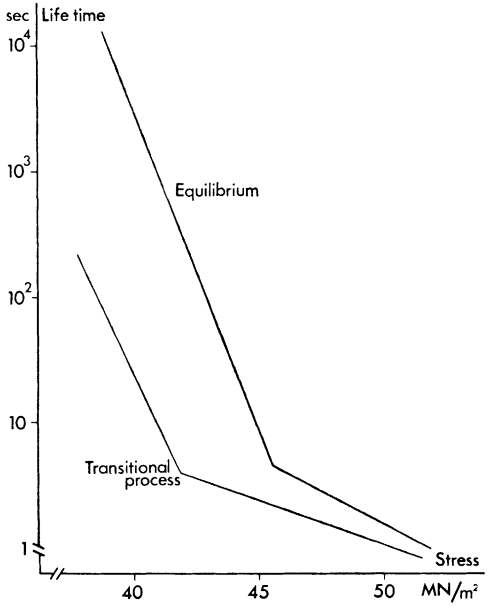

Fig. 18—The creep failure time (lifetime) plotted against stress in tension under equilibrium conditions (65 per cent rh) and transitional conditions (10→65 per cent rh)

Torsional pendulum for single fibres

A SPECIALLY designed torsion pendulum was used to measure the logarithmic decrement and resonance frequency of single pulp fibres. A schematic drawing of this pendulum and the detection system is given in Fig. 20. To record the torsional oscillations, which are electromagnetically initiated, a light beam is focused by means of fibre optics on the borderline between a reflecting and a black zone on the inertia disc. Both the incident and reflected light are transmitted through the same fibre optical system. An oscillatory motion of the disc causes a variation in the reflected light, which for small displacements is proportional to the rotational movements of the disc. A photodiode is used to measure the intensity of the light and the signal is recorded on a conventional potentiometric recorder. The light source is mounted on a turntable, which makes it possible to restore the original intensity by rotation of the optical system. Hence, an absolute value of the final angular rotation of the inertia disc can be obtained. Exponentially decaying damping curves are obtained, from which the logarithmic decrement is evaluated.

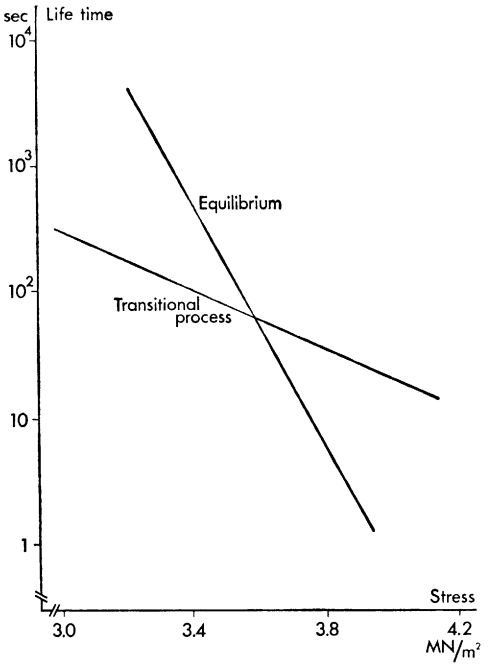

Fig. 19—The creep failure time (lifetime) plotted against stress in compression under equilibrium conditions (65 per cent rh) and transitional conditions (10→65 per cent rh)

The square of the frequency of the free oscillations is proportional to the torsional rigidity of the sample. The theoretical background of the experimental techniques involving free oscillations is thoroughly described in most textbooks of polymer physics.

Experiments were performed in a nitrogen atmosphere or a suitably conditioned air stream. Through the use of a thermostatic waterbath and two heat exchangers, temperature and humidity could be independently controlled in the intervals 10–90° C and 5–59 per cent rh. The weight of the pendulum could be varied 0·2–2 g. The fibres were mounted with a free span of 1 mm according to a technique previously described by Kull.[32] A more detailed description of the torsional pendulum for single pulp fibres appears elsewhere. At room temperatures, a carboxymethylcellulose glue (Modocoll) was used to anchor the fibres; at high temperatures, a resorcinol-based cement was found to be suitable. Epoxy was found to be highly unsuitable because of the penetration of the glue inside the fibres.

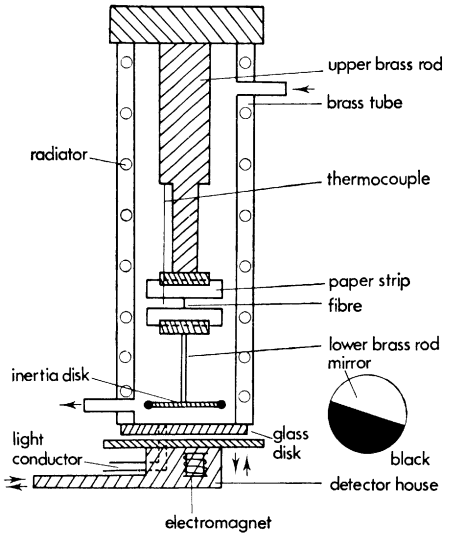

Fig. 20—Schematic drawing of torsional pendulum for single fibres

Torsional pendulum for paper

THE torsional properties of paper were evaluated with a commercial pendulum (Torsional Braid Analyser (TBA), Chemical Instrument Co.). The principle of this testing technique has been previously described in detail.[33]

The internal loss factor (or logarithmic decrement) and the resonance frequency were evaluated for all samples as a function of temperature in a nitrogen atmosphere (flow rate 20 ml/min) and at a temperature scanning rate of 1° C/min. The resonance frequency of the free oscillations was within 0·1–1 Hz. The weight of the pendulum was 42 g and the samples had a width of 0·5 cm and a length of 20 cm. All damping curves were directly analysed by a computer by means of an analog-digital converter connected to the output of the torsional pendulum. Internal loss factor and the square of the frequency of the free oscillations were evaluated by a computer by means of a least-square technique and the results were automatically plotted as the mechanical parameters against temperature.[34]

Moisture regain

A CAHN electrobalance was used to measure moisture regain and the rate of moisture sorption or desorption The balance was put in a thermostatic box. To ensure constant temperature, the samples were placed inside glass tubing submerged in a thermostatic bath. Appropriate salt solutions were used to

regulate the humidity. By changing the box and bath temperature, the temperature could be regulated between 25° and 75° C and the relative humidity between 0 and 90 per cent rh.

Calculations of the shear modulus of papers and single fibres

DETERMINATION of the shear modulus of paper by means of torsional pendulum has been thoroughly discussed by Lindbergson & Kubát.[35] Their method of evaluation was used, but no specific consideration was given to the influence of the weight of the pendulum, since interest was focused in this study on the relative changes of the elastic properties with temperature and humidity. From Kubát's study, a correction factor of 1·1 for calculation of shear modulus was found to be applicable for the present set-up.

The shear modulus of single fibres was calculated from estimates of their cross-sectional area from micrographs of microtomed sections. These sections had a thickness of 15 μm and were cut at several positions along the length of the fibres for which the resonance frequency had been determined. Before microtoming, the fibre was imbedded in methylmethacrylate and dyed in order to yield good phase contrast. The fibres used for the calculation of shear modulus were latewood fibres, thus had thick fibre walls. From the micrographs, it was deemed that the influence of lumen on the moment of inertia was small. The photographs also yielded necessary information about the shape of the cross-sectional area for the calculating of the moment of inertia.

The following formula given by Nielsen[36] was used to calculate the share modulus—

$$G = \frac{630IL\omega^2}{bd^3\mu}$$

where G = shear modulus, dyne/cm^2
I = polar moment of inertia of the oscillating system in g/cm^2
L = length of specimen between clamps in cm
b = width of specimen in cm
d = thickness of specimen in cm
ω = resonance frequency, Hz
μ = shape factor depending upon the ratio of the width to thickness of the specimen[36]

The change in rigidity was taken as the relative change of the square of the frequency.

In calculating the shear modulus of the single fibre at various humidities, it is necessary to compensate for the expansion of the cell wall. In our case, the correction factors given by Meredith was used.[37]

It is realised that the methods of calculating the shear modulus are crude. The estimates of the cross-sectional areas are never precise and they influence

drastically the absolute value of the shear modulus. Nevertheless, reasonably good agreement with previously reported data for ramie fibres[38] was obtained and a good correspondence was found with the theoretically predicted values given by Tang.[39] In our investigation, the mean value of G of three different fibres is given.

Elastic modulus

THE elastic moduli of the samples were determined by means of a conventional tensile tester (Instron). Creep failure times in tension and compression were determined with specially designed equipments similar to those described by Byrd[30] and Guthrie.[40] The environment was controlled by circulating air around the samples. The air was conditioned by recirculation through glycerine solutions of various water contents as described by Kubát.[41]

Dimensional instability

THE hygroexpansion of the sheets was measured by means of a commercial apparatus.* The humidity of the air was controlled by recirculating air through glycerine solutions of water. All sheets were destressed before measurements by subjecting the sheets to 35 per cent and 95 per cent rh for 7 days.

Sheetmaking

THE Scan method was used in the sheetmaking procedure for ordinary handsheets. Oriented laboratory sheets were made by means of a special sheetformer (Formette Dynamique).

Raw materials

FOR the single fibre studies, the wood was handchipped and springwood and summerwood were digested separately.

Characteristic data for the pulps used in this investigation is given in Table 4.

TABLE 4—CHARACTERISTIC DATA FOR THE PULPS USED

Pulp	*Yield, per cent*	*Lignin, per cent*	*Cellulose, per cent*	*Hemicellulose, per cent*
Unbleached sulphate	54	9·0	77·0	14·0
Unbleached sulphate	47	4·5	77·0	18·5
Bleached sulphate	44	—	87·8	11·8
Unbleached sulphite	58	11·4	71·4	15·0
Bleached sulphite	44	—	81·0	18·5

Fibrillar angle

THE microfibrillar angle was measured for some of the fibres used in this investigation. The measurements were made by the use of X-ray diffraction

* Lorentzen-Wettres

and polarised light. In the first case, the measurements were carried out by J. Kratky; in the second case, by D. H. Page. Using the polarised light technique, it was shown that the fibres display a considerable heterogeneity in the orientation of the microfibrillar angle. This is illustrated by the frequency curves in Fig. 21.

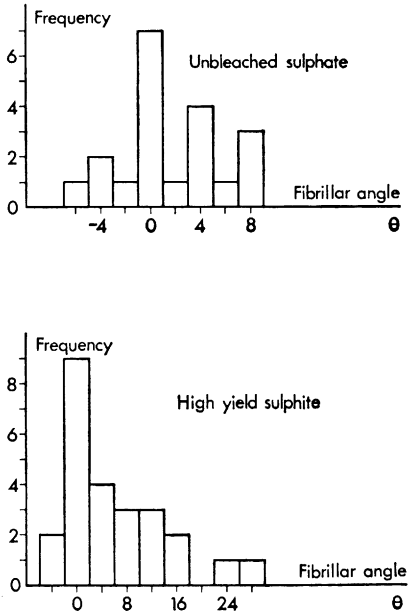

Fig. 21—The variation of fibrillar angle for two different fibre populations

Acknowledgement

The assistance of D. H. Page in making the polarised light measurements is gratefully acknowledged.

Special thanks are also directed to Prof. D. Wahren for valuable criticism of this paper.

Furthermore, a special mention should be given to S. Cavlin, who has made available his work on equality between the geometric mean of the elastic properties of oriented and isotropic sheets.

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Discussion

Mr V. Balodis I would like to comment on a practical aspect of the influence of moisture content and temperature changes on viscoelastic properties of corrugated fibreboard containers.

A few years ago, I was investigating the causes of damage to Australian apple export packs during transport from Hobart, Tasmania to the markets in England. It was generally assumed before the survey that damage to packs is caused primarily by the high loads they have to support in the deep holds on the ship. The survey revealed that relatively few packs were deformed as a result of being shipped in deep holds, but that they were damaged during loading and unloading operations and were most *severely deformed* during the first day after being unloaded from the refrigerated holds on to the wharf and stored under relatively light load.* The sudden deformation can be explained in terms of creep behaviour of cellulose materials, for which creep rate is accelerated at high humidity and during change in humidity and temperature. After unloading, the moisture content of the corrugated board increased on the average from 13 to 16 per cent owing to condensation of water on the cold packs and fruit. Consequently, the deformation of packs was not so much due to the lack of dry compression strength, but to the lack of creep resistance of the board during this critical period. A further significant deformation of the packs took place during the drying out period of the corrugated board.

A practical aspect of the observation is that more attempts should be made to simulate field conditions in the testing laboratory. For example, the packs (or the corrugated board used in their manufacture) should be subjected to suitable atmospheric humidity and temperature cycles while under appropriate load. We have derived suitable laboratory conditions for the testing of export apple packs for tropical markets (such as Singapore and Hong Kong) and markets such as England.† Other test procedures could be designed for other transport and climatic conditions.

* Balodis, V. and Hawkins, B. T., *Appita*, 1972, **25** (5), 362–369
† Balodis, V., *Australian Packaging*, 1971, **19** (2), 22–24

Under the chairmanship of Prof. V. T. Stannett

Dr A. de Ruvo I agree with you that one should at least be aware of these things and to test at equilibrium conditions might not be right for the conditions that the paper is exposed to during handling. You have made a very important point.

Mr P. T. Herdman In several of the graphs, you were showing one of the axes as the elastic modulus of paper. How do you manage to measure the void volume in determining the elastic modulus?

Dr de Ruvo We measured the density by an ordinary caliper measurement. Elastic properties were determined by dynamic and quasi-static techniques. It is a problem that we cannot measure thickness very accurately in our work. I do think we should put a lot more emphasis on this problem than we have done in the past. It is an important problem, but we have done what we could do in the laboratory at this moment.

Mr K. Ebeling The illustration (Fig. V) is shown to explain the fundamental material property of an approximate relationship between the linear thermal expansion coefficient and the extension modulus.

The potential energy curve for two atoms and the related repulsive and attractive force curves are illustrated. The mean separation between atoms is determined by the balance of the opposing forces. Corresponding to this mean separation is the minimum of the potential energy curve.

The slope of the net force plotted against interatomic distance gives a figure related to the extension modulus of the material. The slope is related to the second derivative of the potential energy curve.

As the temperature of the material increases, the mean separation between the vibrating atoms increases. This is shown by the bisecting line of the potential energy curve.

The deeper the 'well' in the potential energy curve, the smaller the linear thermal expansion coefficient of the material. At the same time, the extension modulus of the material—related to the second derivative of the potential curve—will be high. The deeper the well, the higher the second derivative—that is, the higher the extension modulus.

The same result can also be derived by quantum mechanics: Barker* has studied a great variety of materials and obtained that—

$$Y \sim C/\alpha^2$$

where C is a constant. Cellular materials, however, did not follow this relationship completely.

* Barker, Jnr., R. E., *J. appl. Phys.*, 1963, **34** (1), 107–116

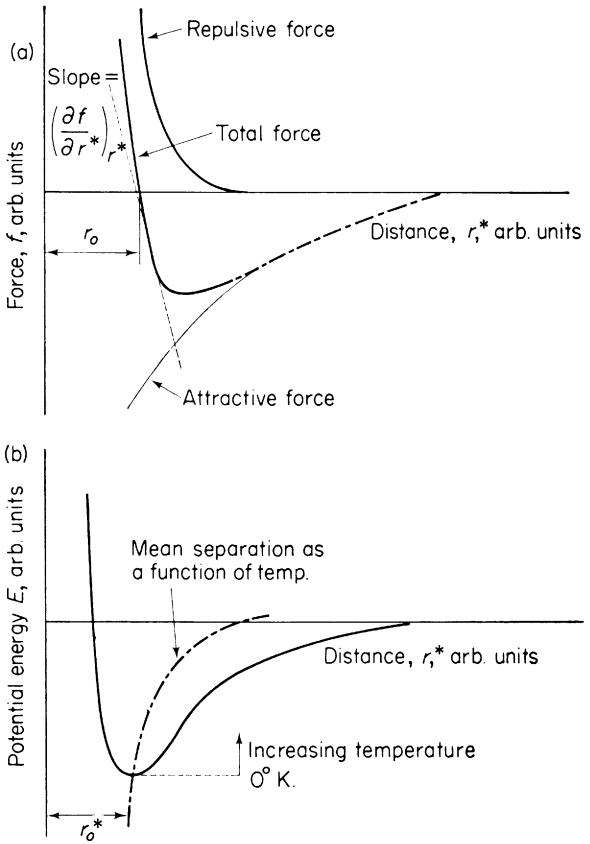

Fig. V—Schematic representation of the force and energy of atomic interaction as a function of interatomic distance

Dr de Ruvo We also have read Barker's paper. It is to be noted, however, that the theory derived by Barker can be applied only to isotropic and homogeneous materials. Especially oriented and heterogeneous materials display a deviation from the general relationship given by Barker.

Mr Ebeling I would like to check a few things from the experimental procedure. How did you measure the thickness of the relaxed sheets? In your paper, you talk about destressing and that a constant value was obtained for the product of the extension modulus times the thermal expansion coefficient. What I am driving at is that, if you measured the thickness of the relaxed

destressed sheet by a simple caliper method, you were probably overestimating the thickness and that would cause an error in the modulus. This would mean that, in relaxing the stressed sheet, the points you show now to be falling on the line would not be falling on the line.

Dr de Ruvo It was a long time ago that we did those measurements, but as I said we used ordinary caliper methods. We know now that we can introduce errors in this way, though it is difficult for me to say what those errors were at that time.

Mr Ebeling The second question has to do with the reported independence of the thermal expansion coefficient from refining. Did you allow the sheets to shrink in a semi-controlled way as they do in the SCAN procedure for handsheet making?

Dr de Ruvo The sheets were made by ordinary forming procedures, but after that we subjected them to variation in climatic conditions. We changed the humidity for 14 days from 95 to 30 per cent. Doing that, we at least believe that we have obtained a destressed sheet. This is very important, as the amount of internal stresses depend on the level of beating or pressing before drying. Very often, therefore, it seems that the mechanical properties of paper are determined on sheets for which the amount of internal stresses is not known. In our case, this source of error was reduced by the destressing operation.

Mr Ebeling Was the modulus measured in dry nitrogen conditions as the thermal expansion was measured?

Dr de Ruvo Yes, they were measured in dry conditions.