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Effects of humidity changes on damping and stress relaxation in wood

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Mechanical damping (tan δ) and stress relaxation on pine veneer samples under stepwise humidity changes were investigated. The loss factor shows a transient peak every time the relative humidity (RH) of the surrounding atmosphere is changed. The effect appears to be associated with the diffusion of water molecules into or out of the material, the peaks being observed both when the sample is humidified (5%–85% RH) or subjected to drying. The results are supplemented by stress relaxation data obtained on similar specimens. Also in this case a higher relaxation rate is observed when the RH level around the sample is changed, the total stress decrease after several cycles being significantly larger than the corresponding value observed after the same period at the higher RH level.

1. Introduction

Despite the fact that wood structures are exposed during normal use to conditions of varying humidity, the dramatic effects of periodic humidity changes on the time-dependent mechanical properties of wood have been reported only rather recently. Armstrong and Kingston [1] found a significant increase in the creep rate of samples of some *Eucalyptus* and *Pinus* samples subjected to bending loads when the humidity of the surrounding atmosphere was periodically changed. Armstrong and Christensen confirmed the effect for thin samples, thus reducing the possible significance of humidity gradients [2, 3]. The rate of change of the deformation appeared to be related to the rate at which the humidity was varied [4]; loading mode, sample orientation, amplitude of the humidity cycles, and average humidity of the sample were some of the variables studied. As expected, considering the high degree of anisotropy of wood, certain differences in the creep enhancement effect were found [4].

The basic findings relating to the increase in creep rate of wood under periodically changing humidity of the surrounding atmosphere have been corroborated in a significant number of papers. Hearmon and Paton [5] found that the Young's modulus of the samples used in Armstrong and co-workers work [1–3] did not change during accelerated creep. Several papers confirm the finding that the creep rate reaches its highest values during desorption [5, 6]. On the whole, the explanations proposed hitherto do not seem convincing. This is especially true of the attempts linking the effects observed with specific features of the wood structure, because similar effects have been observed with a number of other hygroscopic materials, including paper [7, 8] and fibre and particle board [9, 10]. A recent paper by Wang and Dillard [11] gives a number of additional references to textile fibres, paper, and polymers, mentioning transients in mechanical damping during sorption or desorption of humidity or other swelling agents. Wang and Dillard [11] also presented measurements of the mechano-sorptive effect on aramide fibre-reinforced epoxy-based composites. With polyamide-6,6, Hunt and Darlington [12] were unable to find any anomalous creep effect. Among further work, we mention the reports of Hunt [13] and Hunt and Shelton [14] following an engineer's approach to the design of loaded wood structures, and also trying to correlate the effects observed with the structural features of the wood fibres (fibrillar angles, etc.) [15]. Further references may be found in the reviews by Grossman [16], and Wang *et al.* [17], enumerating also the important features of accelerated creep as, for instance, the fact that the creep deformation is independent of the rate at which the humidity is changed.

Several models have been proposed to describe the mechanisms underlying mechano-sorptive effects. Some of the descriptions rely on conventional rheological models supplemented with a friction (ratchet) element [18, 19]. Bazant [20] assumes a Maxwell model to possess a humidity-dependent viscosity. The friction mechanism is, in some cases, given a physical significance of certain structural elements sliding against each other (fibrills, fibre layers, etc.) [19]. Ranta-Maunus [21] treats the effects encountered in analogy with the corresponding phenomena of thermo-viscoelasticity, arriving at rather complicated non-linear integral equations. Hunt [22] attempts a

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qualitative explanation, starting from Barkas' theory of stress-dependent swelling [23].

The aim of the present communication is to supplement the experimental material on the mechano-sorptive effect with data obtained in dynamic measurements on wood (veneer) samples under periodically changing humidity of the surrounding atmosphere. It will be shown that the mechanical damping (loss factor) exhibits pronounced transient peaks when the humidity is changed, irrespective of the direction of this change. Also, the corresponding change of the force to deform the sample (bending mode) will be shown. In addition, some data relating to the acceleration of the stress relaxation process in wood by humidity changes will be reproduced.

2. Experimental procedure

The samples used in the main part of this investigation were obtained from radially cut splint veneer of Scots pine (*Pinus silvestris*). The dimensions of the samples used in the dynamic measurements were 50 mm $\times$ 5 mm. Their thickness was 0.8 and 1.3 mm. In the stress relaxation experiments, the effective sample dimensions were 100 mm $\times$ 6 mm $\times$ 0.8 mm and 100 mm $\times$ 3 mm $\times$ 0.8 mm. The distance between the growth rings was 1.4 $\pm$ 0.1 mm, the proportion of latewood 24% $\pm$ 3%, the density 0.49 g cm^{-3} at 50% relative humidity (RH), and the humidity content 8.4% at that RH level. The sample orientation was longitudinal, transverse and 45°. In the dynamic experiments, samples of balsa wood, thickness 2.7 mm, were also used.

The dynamic mechanical measurements were carried out using a Rheometrics Solids Analyzer RSA II operated in the bending mode. The sample was clamped at both ends, the bending being brought about by an actuator dual cantilever beam attached to the sample middle point. At the beginning of the measurement, the actuator, which also functioned as a force sensor, was adjusted so that the sample was stress-free (no initial bending). Because the clamps, to which the sample ends were attached, were freely movable in the longitudinal direction, the changes in sample dimensions due to swelling or shrinkage were absorbed by the movement of the clamps, thus eliminating unwanted warping effects. Details of the clamp arrangement are evident from Fig. 1. In all experiments, the frequency was 1 Hz. The test amplitude, i.e. the movement of the centre of the specimen, was 0.3 mm in all experiments, corresponding to a longitudinal deformation at the strained side of 0.08%. The lateral strain resolution of the instrument was about 1 μm and the force sensitivity about 0.01 Pa. The chamber enclosing the specimen had an effective volume of 217 ml. The atmospheres used were dry nitrogen gas with 5% RH and humid air, corresponding to 84%–86% RH. Before the measurement, the sample was conditioned for about 10 h in the starting atmosphere. The rate at which nitrogen and the humid air were introduced into the measuring chamber was 25 ml s^{-1}. The air was humidified by passing it, in the form of finely divided bubbles, through two flasks filled with water.

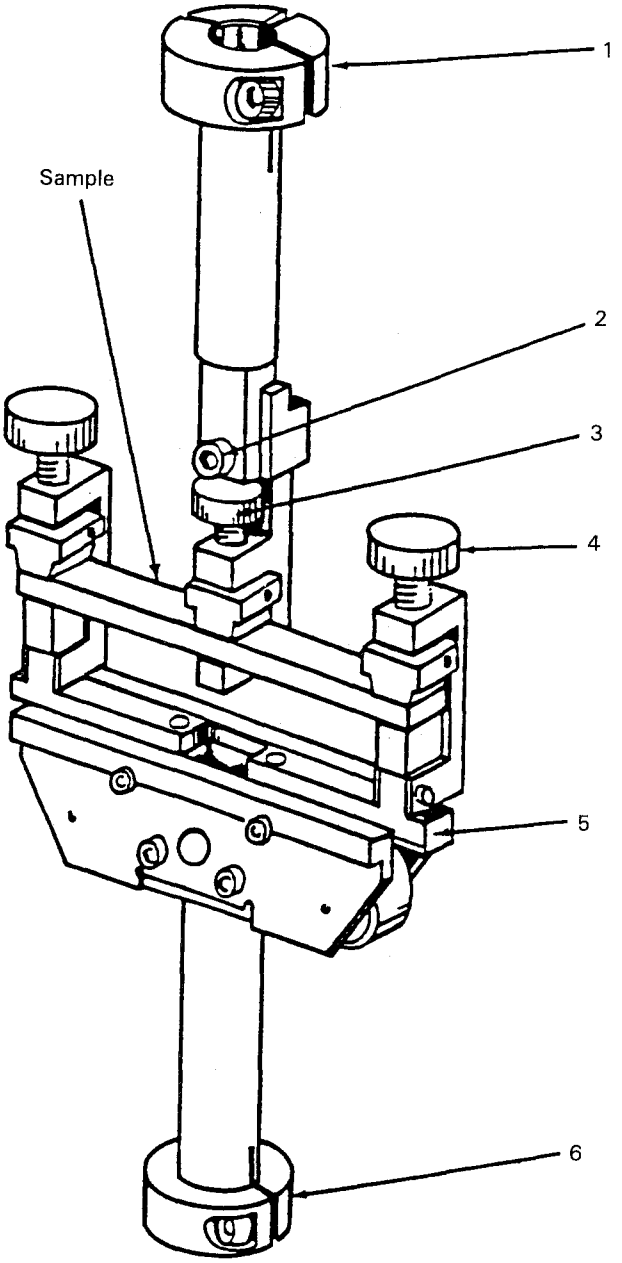

Figure 1 Sample clamping and actuator parts of the measuring equipment (dual cantilever bending fixture, Rheometrics RSA II). 1, 2, 6, set-screws; 3, 4, clamping screws; 5, sliding clamp.

Its humidity was determined as described elsewhere [24, 25].

The equipment used for the measurements of stress relaxation has been described in previous publications [24, 25]. The output from the stress-sensing inductive transducers (Vibro-Meter, TS 1203) was fed into a Vibro-Meter bridge, ICT 617, with a carrier frequency of 8 kHz, and processed in a computer. The sample was contained in a cylindrical enclosure provided with soft rubber membranes having no influence on the recorded stress signal. The volume of the enclosure was approximately 120 cm^3. The required humidity was obtained by circulating air through several flasks with water, thus reaching a near-saturation stage. The flow rate of the humid/dry air was about 3 l min^{-1}. These figures are not considered significant, because the humidity in the enclosure surrounding the sample was recorded simultaneously with the stress, using an electronic hygrometer, Rotronic, type I 128. The rate

at which the atmosphere around the sample changed is shown in the relaxation diagrams reproduced below.

3. Results

3.1. Dynamic mechanical measurements

In the first part of this section we present some typical results obtained in dynamic mechanical measurements on pine veneer samples, showing their response to changes in the humidity of the surrounding atmosphere. The results relate to periodic loading at 1 Hz, with the equivalent longitudinal strain to which the specimens are subjected not exceeding 0.1%. The humidity was changed by blowing dry nitrogen (5% RH) and air with 84%–86% RH periodically into the measuring chamber of the dynamic tester. Fig. 2 shows the variation of the force needed to maintain a prescribed bending amplitude and the mechanical loss factor, tan δ, during the humidity cycling. Starting with the loss factor, one finds that it exhibits a pronounced maximum each time the humidity is changed. This applies both to the drying and humidification part of the cycles. Distinct differences in the shape of the peaks in the two cases are clearly evident, humidification producing peaks significantly sharper than drying. This asymmetry is also reflected in the height of the tan δ maxima. The reproducibility of the data is remarkably high.

When recording the periodic bending force producing a constant amplitude in the middle point of the sample, one observes a different behaviour, in that the force does not exhibit any maxima but varies monotonically upon each humidity change. Upon humidification the force increases rapidly to reach a stage where the rate of increase is very low, while upon drying, the initial rate at which the force decreases is somewhat lower, it continues to fall without reaching a quasi-steady state as observed with humid air. The asymmetry in the behaviour of tan δ is thus also reflected in the variation of the bending force. It may be assumed that this is related to different rates of sorption and desorption.

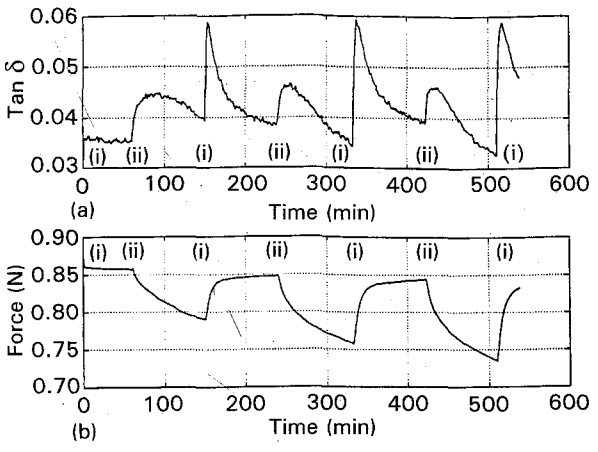

Figure 2 Variation of (a) loss factor, tan δ, and (b) bending force in dynamic mechanical measurements during periodic humidity changes between (i) 85% and (ii) 5% RH. Scots pine veneer, longitudinally cut sample, thickness 0.8 mm. Time period between humidity changes about 90 min.

Gravimetric measurements on small wood samples subjected to similar conditions of humidification and drying exhibit, in fact, small but distinct differences between the two atmospheres. As Fig. 3 shows, the sorption is initially a faster process than is desorption. There is, further, a distinct difference in the rate at which a steady state is approached. However, it is to be noted that the gravimetric measurement relates to minute specimens, only some 10 mg in weight. The correlation with the actual samples used in the mechanical experiments should, therefore, be considered in qualitative terms only.

The data of Fig. 2 relate to experiments where the duration of the dry/humid periods was about 90 min. In the experiments shown in Fig. 4 this period has been significantly extended. As can be seen, the behaviour of tan δ and the bending force are largely similar. Fig. 5 relates to longitudinally cut samples with a thickness of 1,3 mm, i.e. 0.5 mm thicker than those used in the experiments already discussed. As can be seen, this increase in the diffusion path enhances the asymmetry between the peaks corresponding to the two types of atmosphere used.

The measurements reported above relate to specimens cut in the longitudinal direction of the pine veneer. When using transversely cut specimens, an entirely different pattern of the behaviour of tan δ and the bending force is recorded, Fig. 6. While with the

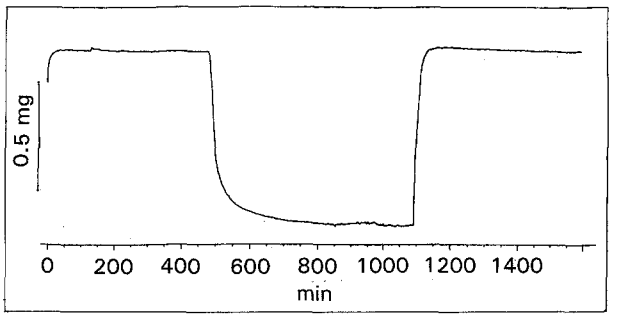

Figure 3 Sorption and desorption curves for a pine veneer sample (about 10 mg) recorded with a TGA device (Mettler M3).

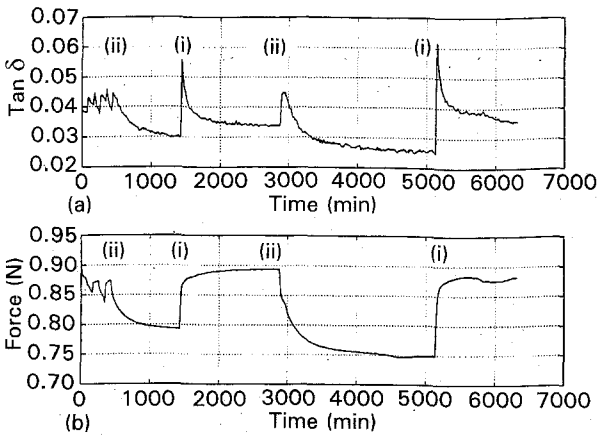

Figure 4 Variation of (a) tan δ and (b) bending force during periodic humidity changes between (i) 85% and (ii) 5% RH. Time period between humidity changes about 900 min. Longitudinal sample, thickness 0.8 mm.

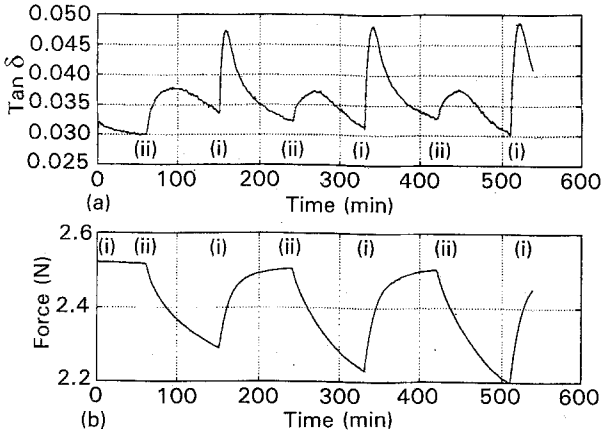

Figure 5 (a) Tan δ and (b) bending force for a longitudinal sample with a thickness of 1.3 mm. Time period between humidity changes ((i) 80%, (ii) 5% RH) about 90 min.

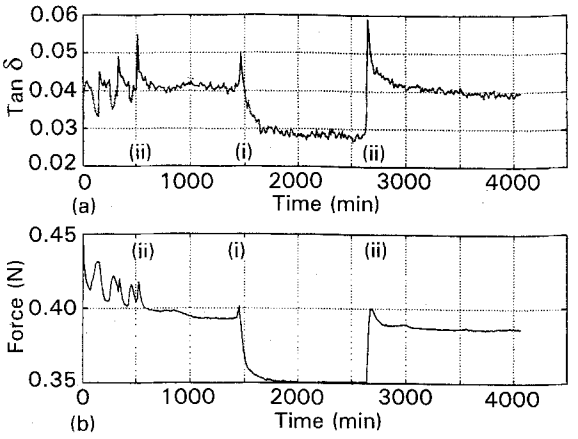

Figure 7 Data as in Figs 5 and 6 for a sample cut in the 45° direction. Period between humidity changes ((i) 85%, (ii) 5% RH) about 900 min.

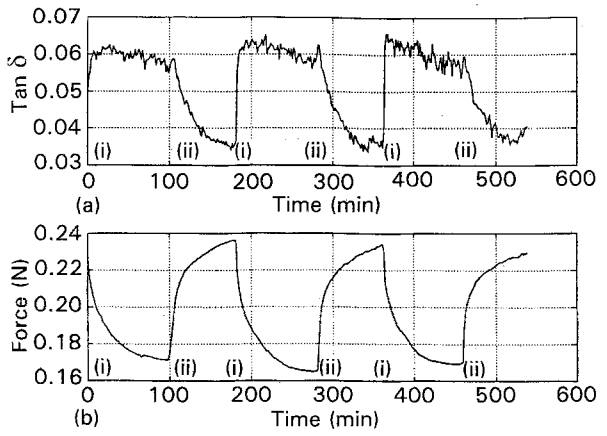

Figure 6 Data as in Fig. 5 for a transversely cut sample of the same thickness.

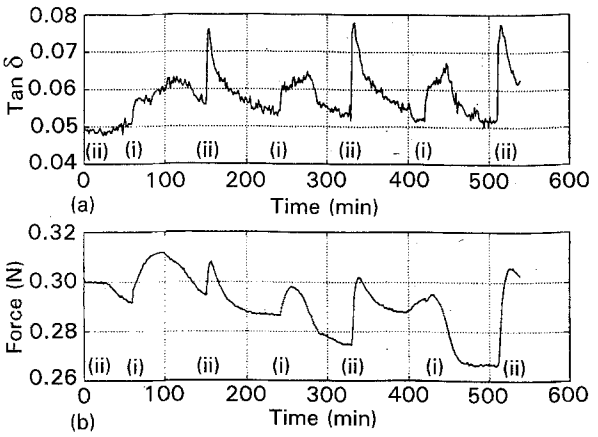

Figure 8 As Fig. 7; period between humidity changes 90 min.

longitudinal samples the force increased upon humidification, and decreased upon drying, one now observed the exact opposite. Also, the damping behaviour is different, the damping levels corresponding to the two atmospheres being reached without any clearly discernible transients. It is to be noted that samples of this type possess an extremely low rigidity, as is evident from the bending force levels in Fig. 6. For this reason only the thicker samples (1.3 mm) could be used in these measurements. The usual thin samples showed visible cracks upon loading, despite the low amplitudes employed. The same applied to samples cut in the 45° direction where, on the other hand, the peaks resulting from a change of the surrounding atmosphere were clearly discernible, although not as pronounced as in the longitudinal direction, Fig. 7. Remarkable, also, was the appearance of minor peaks in the bending force immediately following a change of the atmosphere, an effect not found in the other two directions. As with the longitudinal samples, the bending force decreases when the sample is dried, the opposite being true of the other part of the humidity cycle.

When the period between the humidity changes is reduced, the peaks in the bending force become more pronounced, Fig. 8. On the whole, these peaks coincide with the damping maxima. Apparently, the sharper damping peaks observed upon humidification are accompanied by lower maxima in the bending force.

Fig. 9 may serve as an additional illustration of the effects observed with pine veneer. It reproduces data obtained with longitudinally cut samples of balsa wood (thickness 1.0 mm). As can be seen, the basic features of the variation of the damping and the bending force upon changes of the atmosphere appear also in this case, cf. Fig. 2.

3.2. Stress relaxation

In this section we supplement the transients observed during periodic humidity changes with some data illustrating the effect of such changes on stress relaxation of the pine veneer samples.

The results are presented in the form of stress–time diagrams, also including the variation of the relative humidity as recorded simultaneously in the measuring chamber. The main result of these experiments is the finding that the overall rate of relaxation averaged over the humidity cycles is significantly higher than would correspond to a relaxation process taking place

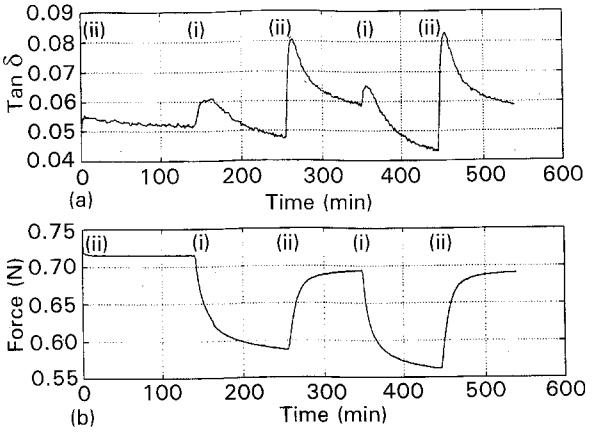

Figure 9 The dynamic mechanical behaviour of a longitudinally cut sample of balsa wood, thickness 1.0 mm.

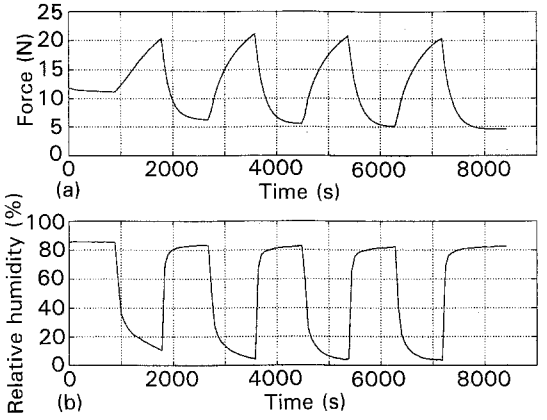

Figure 11 Stress relaxation as in Fig. 10. Humidity cycling with a larger amplitude.

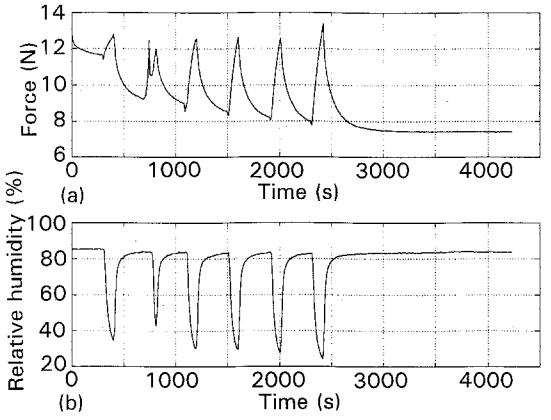

Figure 10 Variation of (a) stress (force) in a stress relaxation experiment with a longitudinally cut Scots pine veneer sample during humidity cycling as recorded in (b). Sample dimensions 100 mm $\times$3 mm$\times$0.8 mm.

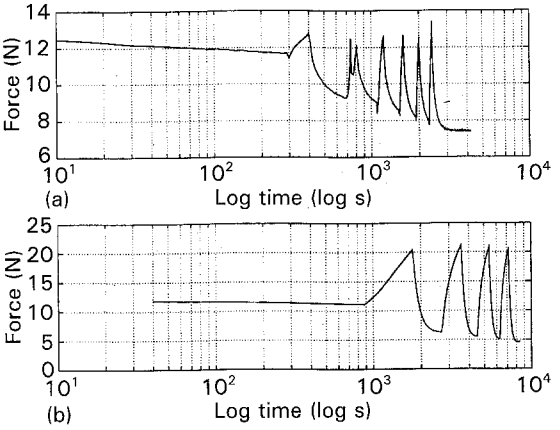

Figure 12 Same data as in Figs 10a and 11b with a logarithmic time axis.

at the humidity level at which the initial loading of the sample took place.

The first example showing the enhancement of the overall relaxation rate is shown in Fig. 10. It relates to 3 mm wide longitudinally-cut samples subjected to a humidity varying between 30% and 85%. For technical reasons, the first loading was performed when the sample was in equilibrium with the higher humidity level. The drying periods were chosen to be somewhat shorter than the opposite ones. Apart from the clearly discernible increase in the overall relaxation rate, it can be seen that the upper peak stress corresponding to the drying stage remains fairly constant, while the humidification periods bring the stress to levels significantly below that to be expected were the initial humidity to be kept constant during the entire process. Based on data obtained earlier [24, 25], a σ/σ_0 ratio (stress/initial stress) of about 0.75 could be expected at 85% RH at the end of the time period covered by the experiment. In the present case, however, we obtain a value of 0.59. Worth mentioning also is the fact that the stress decrease per cycle is largest the first time the humidity is changed, that is, at the beginning of the cycling process.

The results of Fig. 11 relate to humidity cycling between 5% and 85% RH, i.e. a humidity amplitude

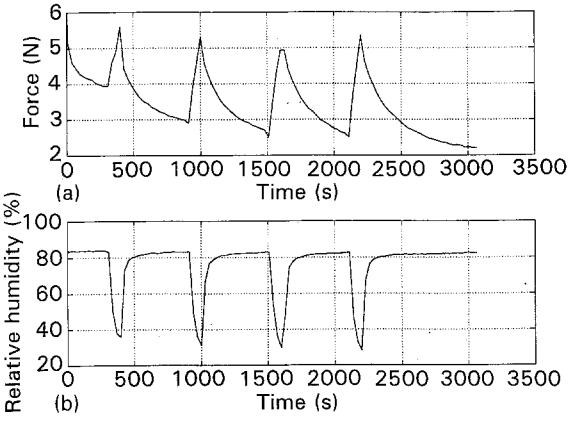

Figure 13 (a) Stress relaxation of a pine veneer sample cut in the 45° direction during humidity changes as recorded in (b).

larger than that referred to in Fig. 10. As expected, this produces correspondingly larger stress amplitudes, the main features of the process remaining similar. This applies both to the constant level of the upper peaks and to the bulk of the stress decrease being concentrated to the first humidity cycle. The final σ/σ_0 ratio reached after about 8000 s is lower than 40%, which is less than the value obtained with the lower humidity amplitude, Fig. 10, and significantly below the level to

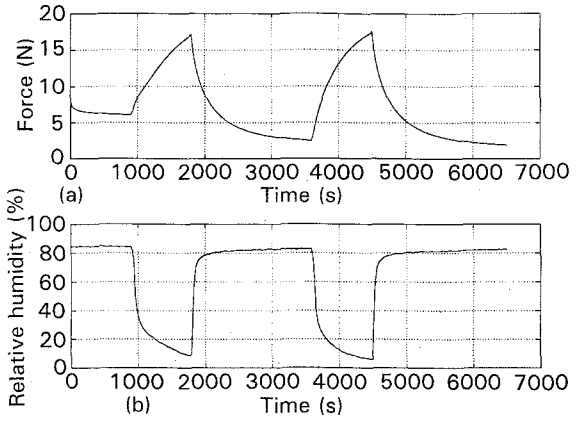

Figure 14 (a) Stress relaxation data for a 45° sample during (b) humidity cycling with a larger amplitude.

be expected at the starting humidity level (marked in the figure).

When plotting σ against linear time, it is difficult, if not impossible, to assess possible indications of an approach to stress equilibrium. In Fig. 12, we therefore show the $\sigma(\log t)$ plots corresponding to the data of Figs 10 and 11. Despite the limited experimental material, the latter data provide a clear indication of a continued fall in stress with the number of humidity cycles.

Pine veneer samples cut in the 45° direction largely behave in a similar fashion. This follows from the results reproduced in Figs 13 and 14, relating to humidity cycles with a relatively low (30%–83% RH) and high (7%–85% RH) humidity amplitude, respectively. Transversely cut samples could not be used in the relaxation experiments, mainly due to their weakness and cracking tendency.

4. Conclusion

Apparently for reasons of experimental simplicity, the bulk of available data on mechano-sorptive effects relates to creep measurements, while other basic modes of viscoelastic behaviour, not so easily amenable to measurement, seem to have remained unexplored. This is the background of the present paper, the purpose of which is to extend the range of experimental techniques to dynamic mechanical behaviour and stress relaxation. As is evident from the results obtained, a periodic change in humidity caused an enhancement of the underlying flow processes in both cases. Apparently, this additional flow, manifesting itself in the damping transients and a higher overall relaxation rate, is due to irreversible movement of water molecules in the loaded wood structure. It appears plausible to assume that such movement, when taking place in a stress field, will dissipate some of the energy stored elastically in the material. Equally plausible is the assumption that, qualitatively, such a dissipation process will be independent of the direction in which the humidity is varied; it may thus be expected to occur both upon drying and humidification.

However obvious such an hypothesis may seem, it has to our knowledge not been proposed when interpreting effects of this type. One reason for this may be the belief that such phenomena occur only in wood, and that a proper explanation has to start from the particular structure of this material [22]. This, however, is not the case as shown, for instance, by the data reported elsewhere [11, 17] and relating to other hygroscopic polymers. Other examples are the damping transients (torsion pendulum) observed during sorption and desorption of water vapour in paper, regenerated cellulose film (cellophane), gelatine, and polyamid-6,6. Similar results were obtained with the systems paper–ammonia, cellulose acetate–acetone, poly(vinyl acetate)–acetone, rubber hydrochloride–chloroform, and polystyrene–benzene [26]. Transients similar to those reported in [26] have also been found, although for torsional rigidity only, with wool fibres [27, 28] upon humidity changes. These effects thus appear to possess a rather general significance extending far beyond the realm of hydrogen-bonded materials. Instead, any molecules moving inside a loaded structure may be expected to produce an interaction between sorption and viscoelasticity, provided such molecules are able to influence the potential field of the solid.

With regard to mechano-sorptive effects during stress relaxation, only some fragmentary experiments appear to have been carried out hitherto. Not unexpectedly, the relaxation rate in bending and torsion of Hinoki wood during sorption was found to increase [29]. Mårtensson [30] changed the humidity in both directions, but the overall effect of the stress level was not clearly evident.

From the physical point of view, the phenomena under discussion are unusually complex. This is certainly the main reason for the unsatisfactory situation regarding their physical interpretation or, in Hunt's words [22], "none of these theories gives more than a partial explanation". For instance, the theory of Barkas [23], although applied in some instances, cannot describe such effects, because it relates to sorption equilibria in stressed solids. In view of the general character of the mechano-sorptive phenomena, and also the recurrence to morphological details of wood structure [22, 31], seems to be devoid of a plausible physical foundation. It may be recalled here that time–dependent processes in solids obey general kinetic laws which are largely independent of the structure and composition of the material under study, cf. for instance Struik [32], relating to physical ageing, stress relaxation and creep.

It thus seems evident that conventional viscoelasticity cannot provide an explanation of these data. This applies also to constitutive equations where the influence of humidity on the various mechanical parameters has been taken into account. Basically, it seems impossible to accommodate the effect within any of the current models of solid state flow. On the other hand, the mechano-sorptive effect offers itself as a novel means to elucidate the nature of the elementary events underlying the flow process, a potential not affordable in equilibrium systems. This, however, requires the design of experiments where a defined number of small molecules moves irreversibly in a

stress field, and where this movement can be interpreted in terms of the energy dissipation which is bound to occur under such conditions. These should be sufficient reasons to refrain, as we do in this communication, from a closer analysis of the experimental results recorded, their main contribution merely being a demonstration of some hitherto unexplored instances of the mechano-sorptive effect. We further refrain from a closer study of the possible role of the loading arrangement used in the dynamic mechanical experiments, such effects being reserved for a forthcoming paper.

Acknowledgements

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References

1. L. D. ARMSTRONG and R. S. T. KINGSTON, *Nature* **185** (1960) 862.
2. L. D. ARMSTRONG and G. N. CHRISTENSEN, *ibid.* **191** (1961) 869.
3. G. N. CHRISTENSEN, *Austral. J. Appl. Sci.* **13** (1962) 242.
4. L. D. ARMSTRONG and R. S. T. KINGSTON, *ibid.* **13** (1962) 257.
5. R. F. S. HEARMON and J. M. PATON, *Forest Products J.* **14** (1964) 357.
6. L. ERIKSSON and B. NORÉN, *Holz Roh u. Werkstoff* **23** (1965) 201.
7. L. BYRD, *Tappi* **55** (1972) 1612.
8. *Idem, ibid.* **55** (1972) 247.
9. L. BYRD and J. W. KONING Jr, *ibid.* **61** (1978) 35.
10. C. BOEHME and C. HARBS, *Holz Roh u. Werkstoff* **42** (1984) 335.
11. J. Z. WANG and D. A. DILLARD, *J. Compos. Mater.* **24** (1990) 994.
12. D. G. HUNT and M. W. DARLINGTON, *Polymer* **21** (1980) 502.
13. D. G. HUNT, *J. Mater. Sci.* **19** (1984) 1456.
14. D. G. HUNT and C. F. SHELTON, *J. Mater. Sci. Lett.* **6** (1987) 353.
15. D. G. HUNT, *J. Mater. Sci.* **21** (1986) 2088.
16. P. U. A. GROSSMAN, *Wood Sci. Technol.* **10** (1976) 163.
17. J. Z. WANG, D. A. DILLARD and F. A. KAMKE, *J. Mater. Sci.* **26** (1991) 5113.
18. R. H. LEICESTER, *Wood Sci. Technol.* **5** (1971) 211.
19. J. MUKUDAI and S. YATA, *ibid.* **20** (1986) 335.
20. Z. P. BAZANT, *ibid.* **19** (1985) 159.
21. A. RANTA-MAUNUS, *ibid.* **9** (1975) 189.
22. D. G. HUNT, *J. Mater. Sci. Lett.* **8** (1989) 1474.
23. W. W. BARKAS, "The Swelling of Wood under Stress" (HMSO, London, 1949).
24. D. G. KUBÁT, S. SAMUELSSON and C. KLASON, *J. Mater. Sci.* **24** (1989) 3541.
25. D. G. KUBÁT and C. KLASON, *ibid.* **26** (1991) 5261.
26. J. KUBÁT and B. LINDBERGSON, *J. Appl. Polym. Sci.* **9** (1965) 2651.
27. B. H. MACKAY and J. G. DOWNES, *J. Appl. Polym. Sci.* **II** (4) (1959) 32.
28. P. NORDON, *Textile Res. J.* **32** (1961) 560.
29. H. URAKAMI and M. FUKUYAMA, *J. Jpn Wood Res. Soc.* **15** (1969) 71.
30. A. MÅRTENSSON, *Wood Sci. Technol.* **22** (1988) 129.
31. P. HOFFMEYER and R. W. DAVIDSON, *ibid.* **23** (1989) 215.
32. L. C. E. STRUIK, in "*Failure of Plastics*", edited by W. Brostow and R. D. Corneliussen (Hanser, New York, 1986) p. 209.

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