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ORIGINAL CONTRIBUTION

Evidence and modelling of physical aging in green wood

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Abstract Physical aging typical of semi-crystalline polymers was evidenced in green (never dried) wood from three tropical hardwood species. The assumption of uniform aging rate was verified by the construction of master curves from series of tensile creep tests in the fibre direction, performed at increasing time elapsed after a quench following heating above the glassy transition. The rheological response during periods of creep small enough to neglect the progress of aging was described by a model made of a spring in series with a parabolic dash-pot where only the characteristic time depends on the aging time. The model was able to describe results obtained by a previous author on softwood loaded transversally to the fibres. The possible role of a transient adsorption process consecutive to the quench is discussed.

Keywords Physical aging · Creep · Green wood

Introduction

As a natural hygroscopic polymer, wood exhibits a complex viscoelastic behaviour depending both on temperature and moisture content. In the living stem, however, the cell cavities contain an excess of free water that keeps the cell walls above moisture saturation. Thus, the rheological behaviour of the building material of a tree, "green" or never dried wood, can be approached by considering essentially the action of temperature, although small variations of bound water content are likely to accompany temperature changes (Skaar 1988).

On the other hand, the process of wood formation involves the production of considerable pre-stressing. Newly formed wood generally contracts after deposition at tree periphery, producing the forces required to control the shape of the stems and enhance their stability and resulting in a well-known field of residual stresses at the macroscopic (stem) level (Archer 1976; Wilson and Archer 1979). This pre-stressing process, however, needs to be considered also at the cellular level. Wood cell wall is a multilayered composite where each layer contains cellulosic reinforcements (microfibrils) oriented at different angles relative to the cell axis and deposited concentrically during cell wall formation (Kollmann and Côté 1968). Cell maturation and death, occurring within few weeks of its life, induce anisotropic deformations of each layer, incompatible to a great extent, so that at the end of the process the cell wall is pre-stressed (Archer 1987). The external stress supported by the wood is modified in the later years when new wood rings have been added to the stem, but a large proportion of these residual stresses and strains remain locked in the cell walls all through the tree life and even after the wood has been cut and processed (Kubler 1987; Gril and Thibaut 1994).

To produce biomechanical models valid over the whole life, the time-dependent behaviour of green

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wood needs to be predicted. The use of series of creep or relaxation tests at increasing temperature levels is the obvious way to identify such long-term rheological models, but several obstacles might produce difficulties. First, the complex polymeric composition might produce overlapping of phenomena so that the observed mechanical response is not described easily by simple equivalence between time and temperature (Bardet and Gril 2002). Second, the recovery of locked-in strain due to the pre-stressing during wood formation at stem periphery is likely to occur during the tests at increasing temperature levels and produce non-negligible perturbation of the viscoelastic data (Gril and Thibaut 1994).

Residual strains can be removed or at least significantly recovered by heating above the glassy transition (T_g), typically 65°C for the in situ saturated lignins of a hardwood (Kelley et al. 1987). However, when the wood is cooled below T_g after the heating, physical aging is likely to occur: Upon cooling below T_g, due to the low chain mobility, the polymer cannot shrink fast enough to keep equilibrium volume, and the material properties undergo slow and gradual changes towards its new equilibrium state (Struik 1978). Consequently, the tests performed too shortly after the heating and subsequent cooling may produce data poorly connected with the real behaviour of a never-heated green wood.

Physical aging has been already observed in wood. A number of arguments for its occurrence were given for wood in the hygroscopic range (Hunt and Gril 1996). The effects of a quench (rapid cooling) on wet wood properties was studied by Nakano (Nakano 2005) who reported a temporary change in viscoelastic properties with a new equilibrium state definitely reached at the end of 10 h. Free volume creation has been ascribed to the freezing of molecular chains of wood components, most likely lignin, during the quench. On the other hand, Ishimaru (2003) has investigated the effect of quenching rate on wood creep properties and observed that the effect of cooling had not completely disappeared after 40 days, but they have not examined the aging phenomenon in more details. In these studies, the wood was usually tested transversally to the fibres, in the radial (R) or tangential (T) direction, whereas for biomechanical consideration, the behaviour in the longitudinal direction (L) is the most important.

The objective of this work was to verify the occurrence of physical aging in green wood, and, if possible, quantify it based on tensile creep tests in L direction. In his pioneer study on physical aging of polymers, Struik proposed an experimental protocol to investigate the effect of aging time on small-strain creep properties after a quench based on the "snapshot" assumption—testing time is very short relative to the aging time. Assuming that aging changes by a negligible amount during each creep test, the creep response can be separated from the aging effect. Struik has also shown that in the case of simple polymers, creep curves for different aging times can be superimposed into a single master curve, and he proposed a simple mathematical description of the log time shift. We adopted Struik procedure, intending to verify the applicability of his descriptive model to green wood.

Experimental

Wood material and creep equipment

Wood samples of three tropical species, *Oxandra asbeckii* (Pulle) R.E. Fries, *Licania alba* (Bernoulli) Cuatrec and *Virola michelii* Heckel, exhibiting contrasting mechanical properties and various anatomical features were used to investigate the diversity of the aging phenomenon in green wood. Plant material coming from tilted trees was included in the study because trees in reorientation process produce particular wood tissues with high levels of pre-stresses and specific cell wall structure (Scurfield 1973), called "tension wood" in hardwoods, unlike normal wood produced a priori by straight growing tree. Sample names are built as the initials of the specie's name with an index specifying the wood type (e.g. Oa_N and Oa_T for *Oxandra asbeckii* normal and tension wood, respectively). Samples (L=150 × T=15 × R=2mm^3) were kept in the wet conditions during all test procedure and storage.

Tensile creep tests were performed on the experimental device used previously by Hunt and Darlington (1978) newly equipped by a water chamber to enable testing of green wood samples (Dlouhá et al., submitted). Water bath was controlled in temperature by a thermo regulator with an accuracy of ±0.1°C. Constant stress was prescribed by assigning an initial displacement of around 500 με corresponding to the generally observed level of pre-stresses in wood tissues. Axial strain in the central part of the sample was measured by a couple of strain gages glued on both side of the sample. Readings were taken every 0.1 s during the first 1,000 s and every second afterwards. We used three-wire gages to avoid the influence of temperature variations on the resistance of the wires and silicon coating to reduce influence of water flow in the bath and risk of gage detachment.

Experimental procedure

After being heated at 80°C for 30 min to release all macroscopic residual strains and establish the thermodynamic equilibrium, the sample was quenched to 30°C and kept at 30 ± 0.1°C. The quench was done by replacing the sample from the bath at 80°C to the water chamber of the testing device regulated at 30°C, the operation taking about 60 s. After that, a set of short-term creep tests was performed on the same specimen at various aging times t_a elapsed after the quench as shown in Fig. 1a. It is important to distinguish aging time t_a, counted from the moment when the specimen was moved from the hot to the cold bath, and t_c, the creep time counted from the moment of each loading. It is always quite delicate to decide which point is to be considered as a starting point of the loading corresponding to $t_c = 0$ s. In this case, the point corresponding to the half load was considered (Fig. 2a). Points corresponding to $t_c < 3$ s were not included in further analysis because of the important dispersion of deformation values attributed to vibrations resulting from the manual weight application (Fig. 2b).

To separate the creep response from the aging effect, testing time t_c has to remain much shorter than t_a at the onset of the test. This is called the snapshot assumption because a test performed during such a short time

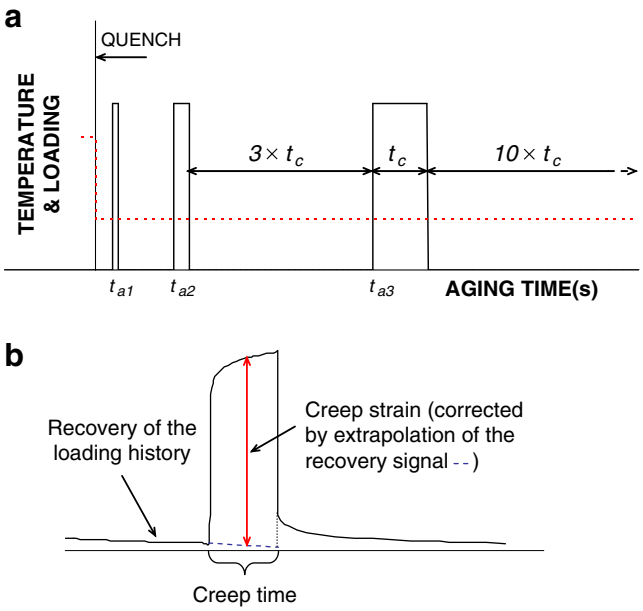

Fig. 1 Experimental procedure for investigating the aging time effect on small strain creep properties. **a** Loading schedule: *dashed line*, temperature; *solid line*, load. t_a aging time, t_c creep time. **b** Accounting for the correction of the creep signal. *Dashed line*, extrapolation of the recovery signal to the creep test duration

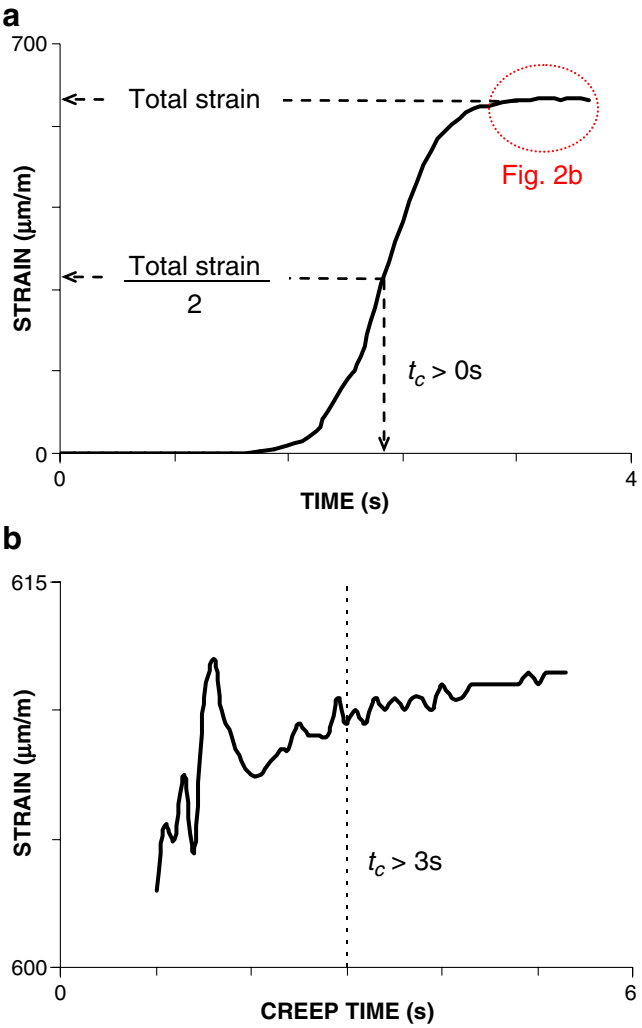

Fig. 2 **a** Method to determine the point considered as the start of the loading. **b** Zoom on the zone of the very first seconds following the loading (unsmoothed data). t_c testing time

($t_c < 0.3 \times t_a$) will exhibit negligible effects of aging. Strain was measured during both creep and recovery periods. By extrapolation of the recovery curve in logarithmic timescale into the next creep period, the strain built up during each individual creep test could be singled out (Fig. 1b). To obtain a reliable extrapolation, testing time has to be short relative to the previous ($3 \times t_c$) recovery period. We also need to have t_c short relative to the following recovery period ($10 \times t_c$). This condition takes into account the time necessary to complete the recovery of previous loading so that at the end strain signal is quite stable and easy to extrapolate. In fact, corrections due to the extrapolation of recovery tests revealed to be negligible (~0.002% for the longest creep test) so that the correction was not applied systematically. The total duration of the test procedure is 3.5 days.

Results and discussion

Suitability of experimental procedure and evidence of aging phenomenon

The experimental procedure was first tested omitting the step of preliminary heating above T_g. In this case, curves corresponding to creep tests performed at different elapsed times after the onset of the test were superimposed into one single curve (Fig. 3a). This shows that the effect of loading history of the sample on a given creep test is negligible and we can use the proposed procedure to investigate the effect of aging time. Figure 3b represents a set of creep tests performed on the same specimen after the quench. We can notice that higher creep strains were developed during the tests following the quench. The relative creep for the creep test performed at t_{a5} (Fig. 3a) amounted to ~7.5% against ~12.2% for the creep test (t_{a5}) in Fig. 3b. The shape of the creep curve corresponding to the first aging time (t_{a1}) was quite different from others. This effect was ascribed to the non-achieved thermal equilibration of the specimen with the surroundings: In later tests, we have always applied the first loading at log $t_{a1} > 2$, or 100 s after the quench.

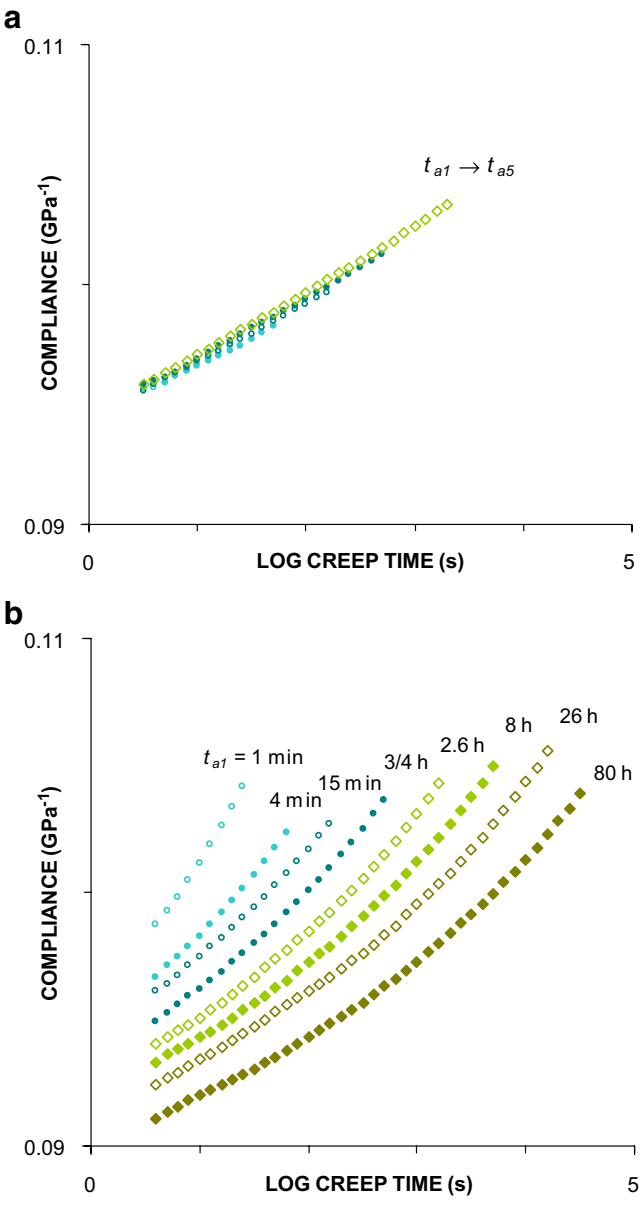

Fig. 3 Evidence of physical ageing occurrence in green wood (sample Vm_N). **a** Set of creep tests performed at different elapsed times t_1–t_5 counting from the onset of the test without preliminary heating above T_g. **b** Set of creep tests performed on the same sample at different aging times t_{a1}–t_{a8} following the quench

The gradual decrease of instantaneous compliance with increasing aging time as visible in Fig. 3b clearly confirms the occurrence of physical aging in green wood. The drop between successive creep curves does not reduce with increasing aging time. This suggests that the logarithmic aging rate remains approximately constant during the test; even at the end of the test period, 3.3 days after the quench, the specimen is far from a new equilibrium state. It definitely shows that the stabilisation period of our samples is much longer that expected considering Nakano's results transversally to the fibres. Figure 4a shows a master curve obtained by shifting the curve for each aging time along the log time axis toward a reference curve corresponding to t_{a8}. The smoothness of the resulting master curve suggests that the aging dependence of the creep response is rather uniform and the material could be considered as rheologically simple. A commonly used method to describe the aging behaviour is to display the shift factor a versus the aging time t_a on double logarithmic scale. The slope yields the aging shift rate μ (Fig. 4b):

$$\mu = -d(\log a)/d(\log t_a)^1. \quad (1)$$

Identification of a rheological model

To improve the reliability of aging quantification, a procedure based on rheological modelling was tried. As a starting point for the model identification, we have used the representation of experimental data in the approximated complex plane (ACP) as proposed by (Huet 1967) for analysis of viscoelastic data obtained by static tests. This method allows checking the presumed validity of creep time–aging time equivalence and offers the possibility to deduce a descriptive model of the phenomenon from the graphical representation

[1] In this paper, decimal logarithms are used.

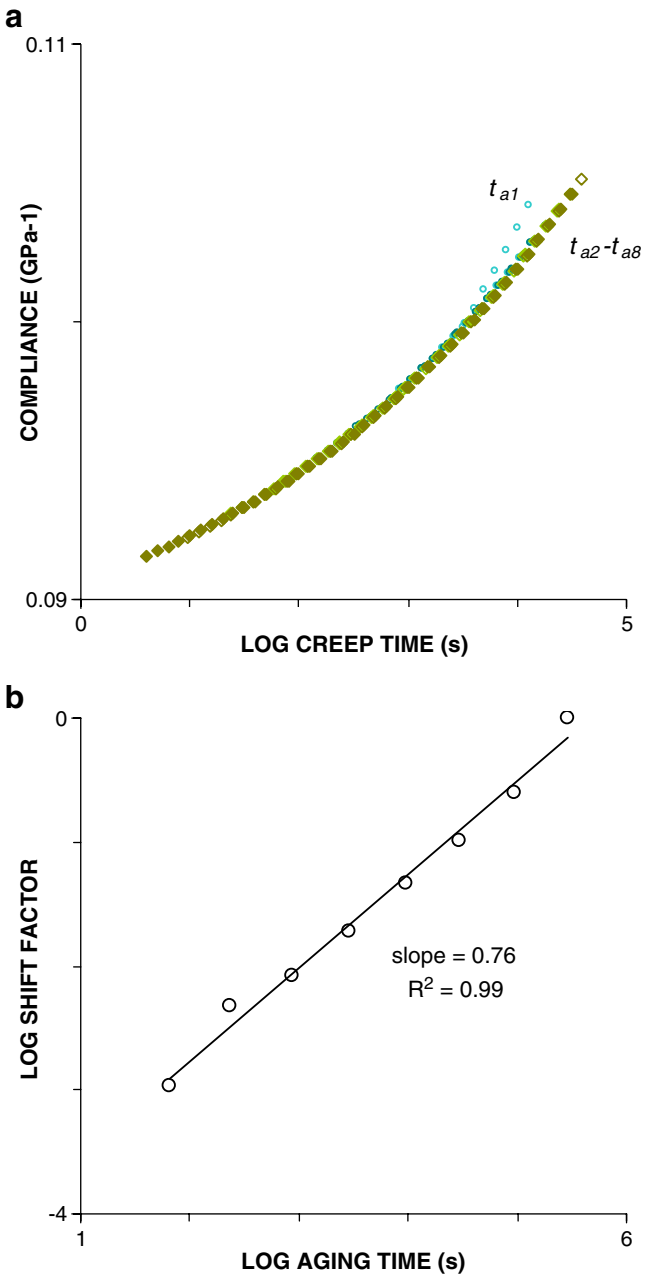

Fig. 4 Relationship between retardation and ageing time based on visual assessment (sample Vm_N). **a** Master curve obtained by visual shifting of individual creep curves from Fig. 3b. **b** Double logarithmic plot of shift factors a versus the aging time t_a

of the data without any presumed hypothesis about its mathematical form. To represent data from static tests, we have used Alfrey's (1948) approximation to obtain storage J' and delayed J'' components of the complex compliance:

$$J' \approx J(t_c)$$
$$J'' \approx \frac{\pi}{2} \times \frac{dJ(t_c)}{d \ln t_c}. \quad (2)$$

The ACP can be understood as a kind of phase diagram showing, for each $J(\log t_c)$ curve, the slope against the value. Alfrey's approximation is valid for small J''/J' ratio and broad viscoelastic spectra, which is usually the case in wood and can be verified a posteriori. In the case of a simple solid rheological behaviour described by a spring in series with a Kelvin block, the representation of the compliance in the ACP deviates significantly from the half circle in the complex plane (CP) used to represent a dynamic response. However, when the dashpot compliance becomes proportional to t^k with $k < 1$, the model is described as "parabolic". For $k << 1$, which corresponds to a broad viscoelastic spectrum, ACP and CP coincide. The applicability of parabolic models to wood was suggested by Huet (1988) and verified for air-dry wood creep in longitudinal direction (Gril et al. 2004) and green wood relaxation transversally to the fibres (Bardet and Gril 2002). The value of k was in the range 0.1–0.5 in these cases.

In complex plot, the creep curves measured at various aging times by form into one single straight line (Fig. 5). This indicates that the aging phenomenon in green wood can be described by a "parabolic Maxwell" model (Huet 1967) with an aging-dependent dashpot element. Creep behaviour of such a model obeys to the power law:

$$J(t_c) = J_0 \left[1 + \left(t_c/\tau\right)^k\right], \quad (3)$$

where $J(t)$ is the creep compliance, J_0 the instantaneous compliance, τ the doubling time of the creep response and k the power parameter. The use of a power law has been often proposed to describe the creep response of wood (Le Govic et al. 1987). This behaviour can be considered as the beginning of a three-element parabolic response as described above for durations considerably smaller than the characteristic time of the dashpot, so that the spring in parallel to the dashpot is subject to a negligible stress.

Estimates of J_0 and k can be obtained graphically as shown in Fig. 5 and have the same value for all aging times. Only τ depends on t_a, so that the creep compliance for any aging time can be written as:

$$J(t_c; t_a) = J_0 \left[1 + \left(t/\tau(t_a)\right)^k\right]. \quad (4)$$

In this equation, the relation $\tau(t_a)$ is not imposed. Assuming, in addition, the validity of aging rate uniformity claimed by Struik (Eq. 1), we obtain:

$$\mu = -d(\log \tau)/d(\log t_a) = -d(\log a)/d(\log t_a), \quad (5)$$

where μ is the aging shift rate where parameter a represents the horizontal shift used in Struik's procedure

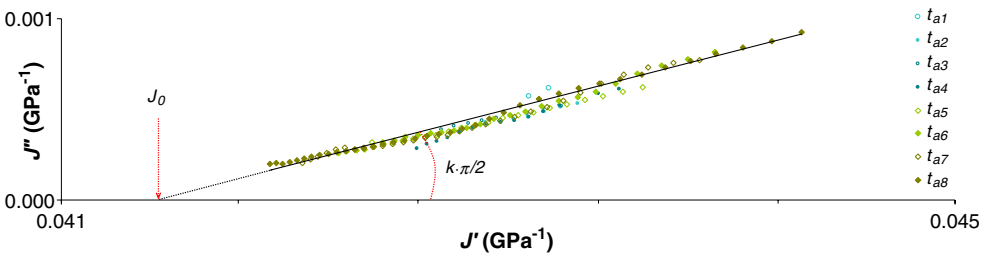

Fig. 5 Complex plot of a sequence of creep tests performed at different aging times t_a after the quench where J' is storage compliance and J'' delayed compliance (sample Oa_N). For signification of J_0 and k, see in the text

to form a master curve from creep curves measured at different aging times.

Equation 5 yields:

$$\log \tau\,(t_a) = \log\,[\tau_0] - \mu\left[\log\,(t_a) - \log\,(t_{a0})\right] \qquad (6)$$

where t_{a0} is the reference aging time and τ_0 the doubling time at t_{a0}. The combination of Eqs. 4 and 6 allows predicting a whole data set like that of Fig. 3b using four parameters only: J_0, k, μ and τ_0. As a reminder, Eq. 4 is only valid for t_c values verifying the snapshot condition ($t_c << t_a$), so that the age of the specimen did not change significantly since it was loaded.

Optimisation of model parameters

Model parameters were adjusted by least-square optimisation method using smoothed experimental data with regular step in log timescale ($\delta \log t_c = 0.1$). First, we have tested the suitability of the proposed description of aging behaviour deduced from the complex plot (Eq. 4). Parameters J_0, k and τ_i were adjusted omitting any presumption about the dependency of τ_i on aging times. This optimisation method will be called "fitting 1". An example of a master curve and adequacy between experimental and modelled creep curves is displayed in Fig. 6. As output of fitting 1, we have obtained a set of doubling times τ_i corresponding to different aging times t_{ai} at which were performed individual creep tests. Plotting the change in τ_i versus aging times t_{ai} in double logarithmic scale (Fig. 6), we can assess the validity of the presumed aging rate uniformity. Excepting the sample Oa_T, the assumption of linearity is well satisfied.

Next, we have proceeded to the optimisation of model parameters J_0, k, τ_0 and μ taking into account both Eqs. 4 and 6, referred to as "fitting 2". An example of master curve using shift factors obtained by fitting 2 is shown in Fig. 8a. We can notice some discrepancies compared to the smooth master curve resulting from fitting 1 represented in Fig. 7a; however, agreement between experimental and modelled creep curves based on parameters issued from fitting 2 remains very satisfactory (Fig. 8b).

Table 1 summarises parameters of the aging-dependent model for all tested specimens adjusted by different methods and/or corresponding to different hypothesis about the doubling times dependency on aging times. Parameter values resulting from fitting methods 1 and 2 are very close. This indicates that the assumption of uniformity of aging rate (Eq. 4) is valid, and so, we can reduce the number of model parameters without introducing an important error to the prediction of aging behaviour. It is also interesting to notice that the accuracy of visual fitting of the master curve is quite good (see values of μ by visual fitting and fitting 1). This study proved that complex plots are very useful in deciding about the mathematical form of the behaviour law; however, the accuracy of value of parameter k deduced from the graphical representation is not sufficient.

Concerning the variability of instantaneous compliances between samples, they can be easily explained by the diversity of densities and Young moduli of our sampling. On the other hand, parameters μ, k and τ_0 were very similar for all samples (excluding aging shift rate μ measured for the sample Oa_T), suggesting the

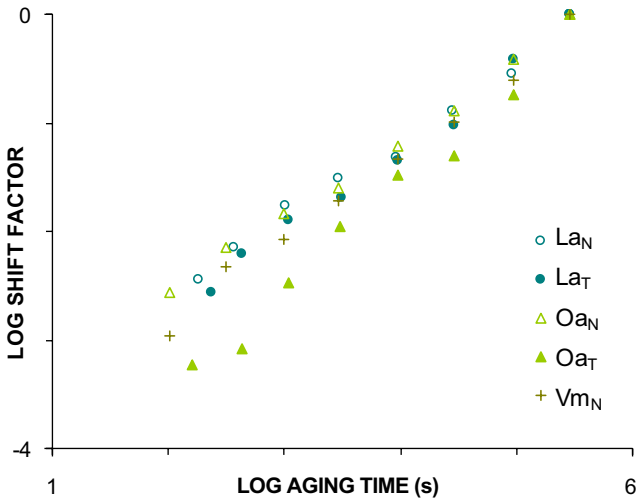

Fig. 6 Model fitting based on Eq. 4. Relationship between shift factors a and aging times t_a in double logarithmic scale for different samples

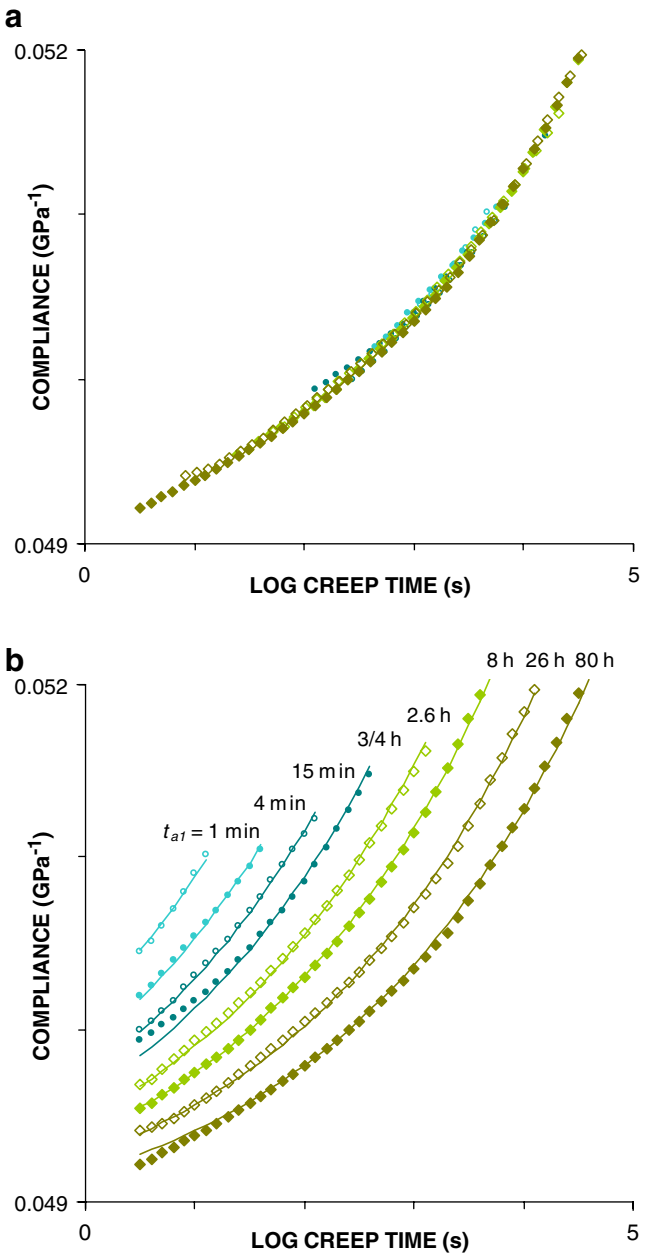

Fig. 7 Example of model fitting based on Eq. 4 for sample Oa_N: **a** Master curve produced by shifts resulting from optimisation without any presumed dependency between doubling and aging times. **b** Example of agreement between experimental (*dots*) and modelled creep curves (*lines*). Creep curve corresponding to the aging time t_{a8} = 3.3 days was taken as a reference

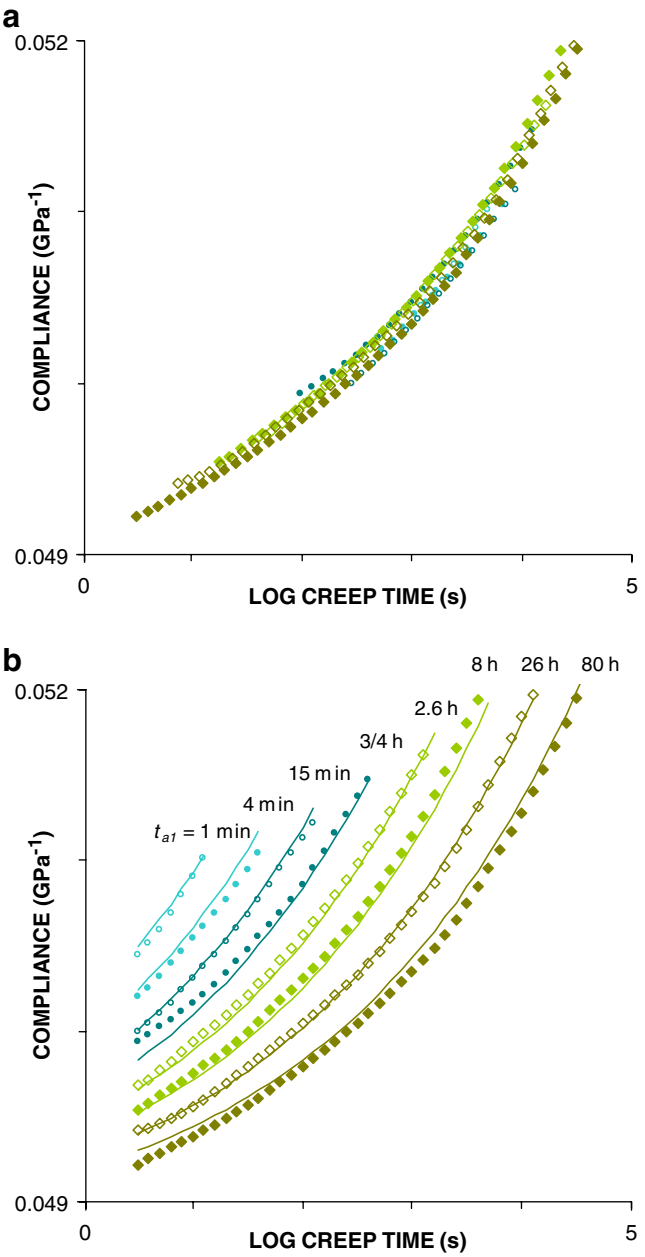

Fig. 8 Example of model fitting based on Eqs. 4 and 5 for sample Oa_N: **a** Master curve produced by shifts resulting from optimisation accounting for linear logarithmic dependency between doubling time and aging time. **b** Example of agreement between experimental and modelled creep curves issued from fitting 2. t_a aging time

common feature of aging phenomenon in green wood. Values of aging rate are consistent with bibliographic data. Struik has observed aging rate μ close to unity for amorphous polymers and $\mu \sim 0.75$ for rigid chain cellulose acetate butyrate ester. Considering partially crystalline and partially amorphous nature of wood, lower aging rate can be explained by the slowing effect of rigid chains as proposed by Levita and Struik (1983).

Application to the analysis of Nakano's data

Nakano studied *Picea jezoensis* (Carrhas), a Japanese softwood, loaded in bending in the *R* direction. Although the wood was studied in the water saturated state, it had been previously kiln-dried so cannot be considered as green wood. The experimental procedure has been different from that suggested by Struik.

Table 1 Adjusted parameters of aging-time-dependant model

	Vis. fitting	Complex plot		Fitting 1				Fitting 2			
	μ	k	J_0	μ^a	k	J_0	$\log\tau_0$	μ	k	J_0	$\log\tau_0$
La_N	0.70	0.21	0.035	0.71	0.17	0.041	11.7	0.72	0.18	0.041	11.4
La_T	0.79	0.12	0.027	0.79	0.14	0.033	12.4	0.80	0.14	0.033	12.4
Oa_N	0.77	0.21	0.042	0.73	0.19	0.048	10.8	0.75	0.19	0.048	10.7
Oa_T	0.98	0.14	0.037	0.97	0.13	0.033	14.2	0.97	0.13	0.033	13.8
Vm_N	0.77	0.14	0.087	0.80	0.14	0.086	9.5	0.80	0.14	0.086	9.3
Nakano				0.73	0.27		5.2	0.61	0.27		5.2

Last line corresponds to results obtained for Nakano's data presented in the second part of the paper, k power exponent, J_0 instantaneous compliance (GPa^{-1}), $\log\tau_0$ doubling time of the reference element, μ the aging rate
[a]Value of μ for fitting 1 is deduced from the plot represented in Fig. 6

Whilst in our case aging times at which individual creep tests are performed are equidistant in logarithmic scale (step = 0.5), in Nakano's procedure, the aging time step is not constant and tends to decrease for longer aging times ($\log t_{an+1} - \log t_{a\,n}$: 0.78, 0.48, 0.30, 0.37, 0.22, 0.27, 0.19). Hence, it is difficult to deduce from the graphical representation of creep curves with respect of testing time if the material is close to its equilibrium state or not.

To examine in more details Nakano's data, we have applied the optimisation method 1—fitting without presumed linear logarithmic dependency of doubling times on aging times. As creep tests were not performed on the same sample, we have allowed J_0 to vary accounting for possible differences in instantaneous and delayed behaviour between samples. Only data fulfilling the snapshot condition $(t_c > t_a/3)$ were used for the fitting. Agreement between experimental and modelled creep curves is shown in Fig. 9a. Relation between shift factors and aging times is quite linear, yielding aging rate equal to 0.73 (Fig. 9b) that is very close to our values for tropical hardwoods loaded in longitudinal tension. It suggests that the dynamics of aging phenomenon is not dependant on the wood type or the loading direction.

Discussion on the physical processes involved

According to Stamm and Loughborough (1935), the fibre saturation point (FSP), representing the maximum content of bound water, increases by 0.1% per 1°C decrease. This estimate, deduced from sorption isotherms, is representative of phenomena occurring in the hygroscopic domain. However, even in wet conditions, with the cellular cavities (lumens) filled with free water and the cell walls saturated with bound water, small moisture movements are likely to occur through the cell wall as a result of temperature changes.

The phenomenon stated by Stamm is qualitatively confirmed by the negative reversible thermal expansion observed in green wood and attributed to a moisture

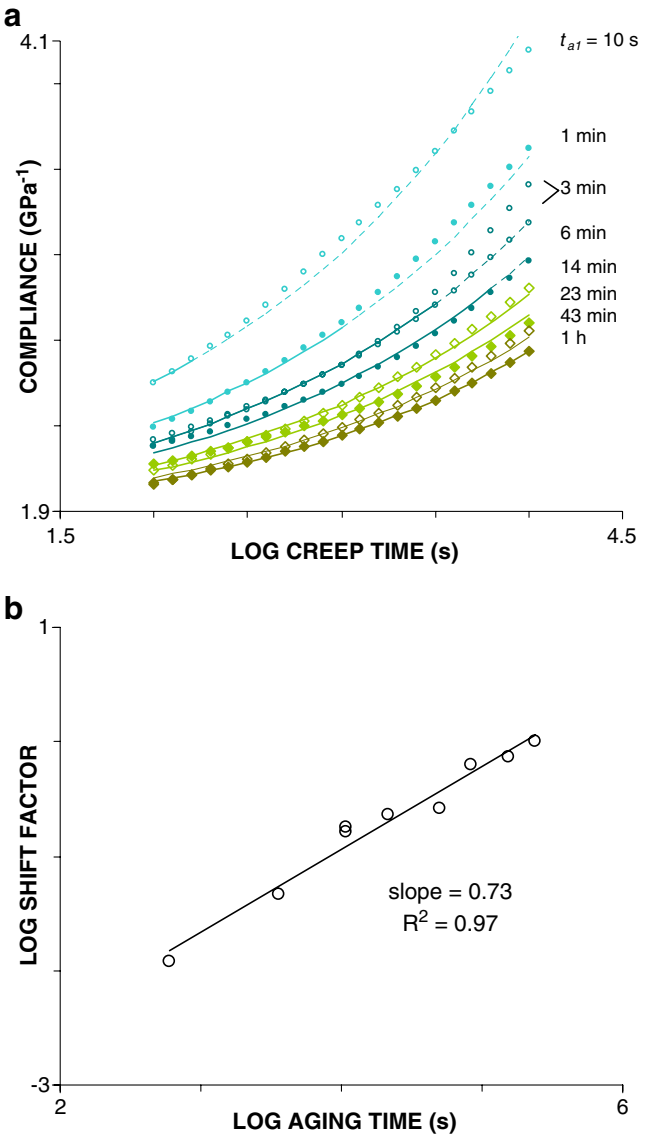

Fig. 9 Model fitting based on Eq. 3 applied on Nakano's data: **a** Agreement between experimental and modelled creep curves for fitting 1. *Solid lines* represent the prediction for values of $t_c < t_a/3$; *dashed lines* correspond to modelled values not satisfying the snapshot condition $t_c > t_a/3$. **b** Relationship between shift factors a and aging times t_a in double logarithmic scale

uptake during cooling (Yokota and Tarkow 1962). But the intensity of the phenomenon seems to be smaller. An expansion ratio of -7.10^{-6}/°C in the tangential direction can be evaluated for green wood from a previous study (Gril et al. 1993). Taking $+33.10^{-6}$/°C as the expansion ratio for dry wood (Kollmann and Côté 1968), the expansion resulting from the moisture uptake can be calculated as -40.10^{-6}/°C. Assuming a typical expansion ratio of 0.3% per percent moisture content, this would correspond to a moisture content increase of 0.7% for a 50°C temperature decrease to be compared to the 5% predicted from FSP estimates.

Even by that small amount, the moisture uptake resulting from quenching is likely to influence the mechanical response. In the hygroscopic range, humidity variations, both adsorption and desorption, considerably accelerate the viscoelastic response, a phenomenon known as mechanosorptive effect (Grossman 1976) and described as a transient increase of molecular mobility (Back et al. 1983) of the hygroscopic matrix made of lignin, hemicelluloses and non-crystalline cellulose. However, at high humidity levels, the contributions of mechanosorptive and stable-state creep are difficult to separate. Our data do not allow distinguishing that "hygrothermomechanical" process from the progressive return to equilibrium of temporary frozen molecular segments. The quenching provoked, for a number of reasons, an unstable molecular configuration of the matrix that is progressively recovered according to the identified kinetics.

The fact that Struik's formalism seemed to be relevant to describe the physical aging of green wood suggests that the pertinent level of description is that of a semi-crystalline polymer where the heating above lignin glassy transition followed by a quench had induced a delayed return to a stable molecular configuration. The transient instability induced by the small moisture uptake does not necessarily contradict this interpretation as long as it only adds a reason for higher molecular mobility. However, if the variation in moisture content was the main cause for the change in behaviour with t_a, the remarkable superimposition of the creep curves in Fig. 5, resulting in a model where only τ varies with t_a, may not have been observed.

Higher levels of structural organisation should be also evoked to complement this molecular interpretation. In the longitudinal direction, because of the axial orientation and tubular shape of most cells, the cell wall level is most appropriate. When a green wood specimen is isolated and heated, stress resulting from the cell maturation process is easily recovered as soon as the temperature exceeds T_g, which was the case at 80°C. However, the condition of structural integrity limits the extent of recovery; a part of the locked-in strains and pre-stresses remain present locally, as their complete relaxation would require the destruction of the cell wall structure. The local swelling anisotropy, and its incompatibility between layers, might interact with this initial pre-stressing resulting from cell wall formation and be only partially released by the heating.

Conclusion

The effect of quench on creep persists for a long time in green wood and could be described as a physical aging process. The analysis of aging behaviour was performed in three steps. First, rough assessment of the validity of the aging rate uniformity was made by visual shifting of individual curves into a master curve. Second, complex plots were used to choose a rheological model and identify its parameters for each aging time t_a without any presumption about the influence of t_a. The model used, a parabolic Maxwell, describes the delayed compliance by a simple power law. Only the "doubling time" theoretically required to double the instantaneous response was affected by t_a (fitting 1). Based on results of this fitting, the validity of the assumption of the uniform aging rate was verified and introduced in the model in a third stage (fitting 2). The same approach was successfully applied on experimental data provided by Nakano. Model based on the linear dependency between doubling and aging times was considered satisfying for all studied samples suggesting common feature of the aging phenomena in wood. The transient adsorption process consecutive to the quench was considered as one of the possible causes of the higher molecular mobility observed.

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References

Alfrey T (1948) Mechanical behaviour of high polymers. Interscience Publisher, New York

Archer RR (1976) On the distribution of tree growth stresses. Part 2: stresses due to asymmetric growth strains. Wood Sci Technol 10(4):293–309

Archer RR (1987) On the origin of growth stresses in trees. Part 1: micro mechanics of the developing cambial cell wall. Wood Sci Technol 21(2):139–154

Back EL, Salmén L et al (1983) Transient effects of moisture sorption on the strength properties of paper and wood-based materials. Sven Papperstidning 86:61–71

Bardet S, Gril J (2002) Modelling the transverse viscoelasticity of green wood using a combination of two parabolic elements. C R Méc 330(8):549–556

Gril J, Thibaut B (1994) Tree mechanics and wood mechanics: relating hygrothermal recovery of green wood to the maturation process. Ann Sci For 51:329–338

Gril J, Thibaut B et al (1993) Recouvrance hygrothermique du bois vert. I. Influence de la température. Cas du jujubier (Ziziphys lotus (L) Lam). Ann Sci For 50:57–70

Gril J, Hunt D et al (2004) Using wood creep data to discuss the contribution of cell-wall reinforcing material. C R Biol 327(9–10):881–888

Grossman PUA (1976) Requirements for a model that exhibits mechano-sorptive behaviour. Wood Sci Technol 10(3):163–168

Huet C (1967) Représentation des modules et complaisances complexes dans les plans complexes arithmétique et logarithmique. Cah Groupe Fr rhéol 5(1):237–258

Huet C (1988) Some aspects of the termo-hygro-viscoelastic behaviour of wood. Colloque Scientifique Européen, Bordeaux

Hunt DG, Darlington MW (1978) Accurate measurement of creep of nylon-6,6 at constant temperature and humidity. Polymer 19(8):977–983

Hunt DG, Gril J (1996) Evidence of a physical ageing phenomenon in wood. J Mater Sci Lett 15:80–82

Ishimaru Y (2003) Mechanical properties of wood in unstable states caused by changes in temperature and/or swelling. In: Proceedings of the second international conference of the European society for wood mechanics. Stockholm, Sweden, 25th–28th May 2003

Kelley SS, Rials TG et al (1987) Relaxation behaviour of the amorphous components of wood. J Mater Sci 22(2):617–624

Kollmann FFP, Côté WA Jr (1968) Principles of wood science and technology, vol 1. Solid wood. Springer, New York

Kubler H (1987) Growth stresses in trees and related wood properties. For Prod Abstr 10(3):62–119

Le Govic C, Felix B et al (1987) Mise en évidence d'une équivalence temps-température et modélisation du fluage du bois 1 Documents généraux–Thèmes 1 et 2. Actes du 2e colloque Sciences et Industries du Bois, Nancy, 22–24 avril 1987–tome 1, Nancy, A.R.BO.LOR

Levita G, Struik LCE (1983) Physical ageing in rigid chain polymers. Polymer 24(8):1071–1074

Nakano T (2005) Effects of quenching on relaxation properties of wet wood. J Wood Sci 51(2):112–117

Scurfield G (1973) Reaction wood: its structure and function. Science 179:647–655

Skaar C (1988) Wood–water relations. Springer, Berlin

Stamm AJ, Loughborough WK (1935) Thermodynamics of the swelling of wood. J Phys Chem 39:121–132

Struik LCE (1978) Physical aging of amorphous polymers and other materials. Elsevier Scientific Publishing Company, Amsterdam

Wilson BF, Archer RR (1979) Tree design: some biological solutions to mechanical problems. Bioscience 9:293–298, May

Yokota T, Tarkow H (1962) Changes in dimension on heating green wood. For Prod J 43:43–45