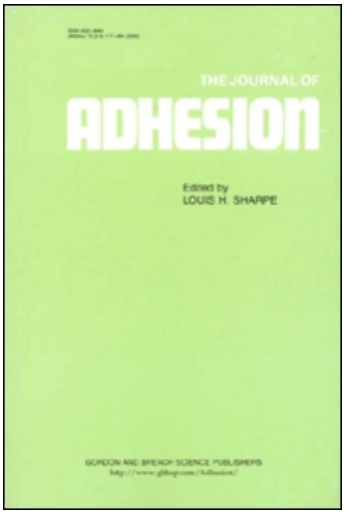

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Predicting Creep Behavior of Wood Adhesives by Torsional Braid Analysis†

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Rheological changes in four cured wood adhesives exposed to temperature and humidity conditions were determined using torsional braid analysis. The adhesives ranged from thermosetting to thermoplastic types. Creep extension in shear also was measured for wood laminates bonded with these adhesives and exposed to conditions of 27°C/90% RH and 60°C/60% RH. A positive trend was found between creep extension and both the maximum rigidity change and the damping loss occurring during humidity and temperature exposure. These findings suggested TBA can be a useful method for predicting wood adhesive creep and failure behavior in the presence of heat and moisture.

INTRODUCTION

The long-term performance of glued wood products is critical to the use of these composites in structural applications. Much effort has been directed toward better understanding adhesive properties and the interaction between adhesives and wood[1,2] in order to predict performance. Presently, the only definitive means of ensuring that a wood bond will

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satisfy exterior service requirements is to conduct long-term test fence exposures. Accelerated methods of wood bond evaluation, involving mechanical testing of gluelines after exposure to moisture and heat, have been used to provide some indication of relative bond durability expectations[3]. With adhesive technology often playing a leading role in product developments additional bond evaluation methods are needed to rapidly establish performance reliability.

Using the premise that the chemical, thermal, oxidative or mositure sensitive properties of the adhesive govern glueline performance, we have applied over the years, various chemical and physical test methods such as thermal analysis,[4] thermal softening[5] and hydrolysis[6] to predict adhesive durability. This battery of test methods yields information which allows for a quick comparison of adhesive cure and performance capabilities. It was recognized that additional techniques which focus on the morphology of the adhesive and its relationship to performance both during and after cure were desirable.

Extensive studies of thermoplastic polymers[7,8] have indicated that useful physical information on material properties can be gained over a short time period using dynamic mechanical methods (DMA). These methods measure the response or deformation, with time, of a material to periodic or varying forces. In particular, one method, torsional braid analysis (TBA), has been utilized to study cure and thermal stability of wood adhesives.[9,10] Since creep behavior of polymers and rheological changes as measured by TBA are based on similar molecular phenomena, it appeared likely that TBA experiments conducted during temperature and/or humidity exposure would offer data useful for predicting relative glueline deformation properties under load. The presence of a loading stress on the glueline is known to influence, over a period of time, the bond durability and load bearing properties of a wood composite.[11]

Creep is an important parameter in structural adhesives and one that can differentiate polymer systems. At present creep response of gluelines usually are determined using long-term, large-scale tests. Thus, any success in relating adhesive TBA response under varying environments to creep response tendencies will aid in the future evaluation of wood adhesive systems for structural considerations.

EXPERIMENTAL

Materials

All adhesive materials were obtained from commercial sources. The phenol-resorcinol formaldehyde (PRF) adhesive (Borden's LT-75) consisted of a two-component liquid prepolymer (50% solids) and powdered hardener system which were mixed just prior to adhesive use. The hardener composition, a 1 : 1 mixture of paraformaldehyde to walnut shell flour, was added at a level of 15 parts to 100 parts resin. The plasticized phenol formaldehyde (PPF) resin (45% solids) was an experimental phenol formaldehyde adhesive containing 20% w/w polyethylene glycol 400. The polyvinyl acetate (PVA) adhesive (Borden's Wonderbond 904) contained a phenolic modifier. The adhesive was a two-component system with the catalyst portion (5 wt/wt percent) being added just prior to use to initiate crosslinking during pressing. In addition, a newer type experimental emulsion polymer isocyanate (EPI) adhesive was evaluated. This adhesive consisted of two parts, a high molecular weight polyvinyl acetate/alcohol emulsion (61% solids) and a polymeric methylene-diphenyl-di-isocyanate component (75% solids) which were mixed together at respective 90 : 10 amounts† based on weight prior to use.

Instrumental methods

Torsional braid analysis (TBA) thermograms were measured on a modified Chemical Instruments TBA[10] at a heating rate of 3°C/min. A multifilament glass braid with cross-sectional diameter of about 0.5 mm and length of 200 mm was impregnated by submerging in a resin solution for 25 minutes. Excess resin was removed by drawing the braid over a paper towel. The braid was clamped onto the TBA sample holders, placed in the oven, and the temperature raised at a programmed heating rate (3°C/min) in a nitrogen atmosphere.

A torsional displacement pulse of 30° was initiated every 75 seconds

† The amount of isocyanate component was chosen to provide a moderately crosslinked EPI. More recent EPI formulations have employed larger isocyanate portions giving correspondingly higher crosslink densities. In these cases any excess isocyanate would likely react directly with the wood and lead to enhanced bond properties. We are presently examining the use of a wood strip in place of the fibreglass braid for the TBA measurement in order to study this situation.

and the braid allowed to oscillate freely until the next pulse. Both relative rigidity ($1/p^2$) and damping loss (Δ) values were determined from each oscillation pattern.[10] For some isothermal TBA measurements at 21°C, a humidity box containing a saturated Na_2CO_3 solution over which air was circulated by a pump (2000 cc/min), was attached to the sample chamber to provide a relative humidity of 92%.

Block shear tests

Block shear specimens were prepared and tested as described in ASTM 905-76[12] except that Douglas-fir blocks were used and the loading rate was 5 mm per minute. Five lamination strips were prepared for each adhesive with PVA, EPI and PRF cured at 60°C for 5 hours and the PPF lamination cured at 120°C for 1.5 hours. Ten dry block shear specimens corresponding to each adhesive set were chosen at random for testing. Shear strength and wood failure were determined and the proportional limit in shear measured from recorded stress-strain curves.

Creep evaluation

Two methods were used to determine relative glueline creep. The first follows procedures described in ASTM D3535-76[13], whereby glued laminations are subjected to constant load under different temperature and humidity conditions and the glueline deformation measured. The Douglas-fir laminated wood samples were prepared as described in D3535-76 with PRF, PVA and EPI adhesives cured at 60°C for 5 hours. The PPF adhesive was cured at 120°C press temperature for 1.5 hours. In all cases 1.05 MPa laminating pressure was used. Samples were allowed to equilibrate at 20°C/60% RH for seven days after pressing. Creep tests were conducted on two samples for each adhesive at 27°C and 90% RH for 7 days and at 71°C and approx. 20% RH for 7 days.

A second creep test method used double shear specimens (Figure 1) similar to those described by Dong and Hoyle.[14] These samples were made with clear Douglas-fir wood with laminations cured as indicated for the ASTM tests above. Four samples were tested for each adhesive condition and an average creep extension curve constructed. Load levels were chosen to be 28% of the proportional limit of the dry shear load deformation curve (Table I). Test conditions were 7 days at 27°C and 90% RH and up to 7 days at 60°C and 60% RH. Glueline deformation was measured every 15 minutes for the first two hours then at one

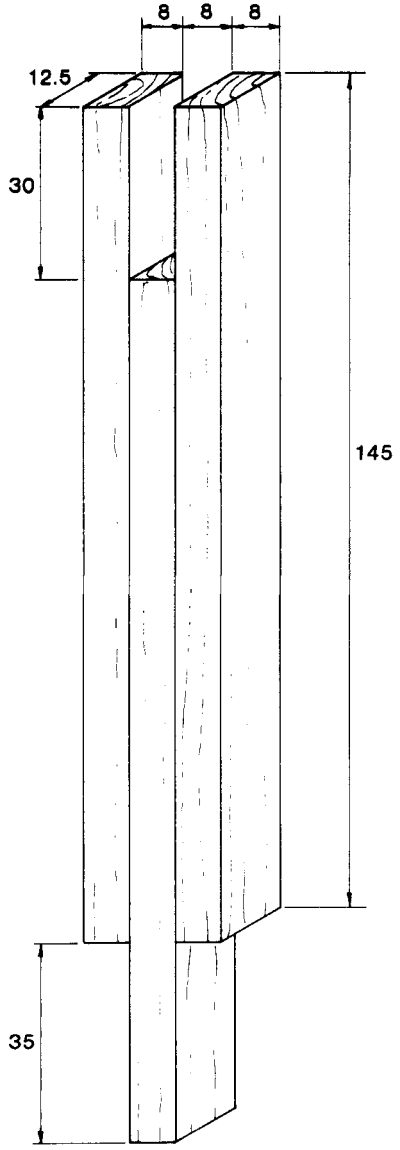

FIGURE 1 Double shear specimens for creep tests.

TABLE I
Dry block shear test results† for adhesive laminates

Adhesive	Shear strength (psi)	Wood failure (%)	Proportional Limit (psi)
PRF	1665	83	700
PPF	1674	95	638
PVA	1670	94	607
EPI	1374	95	645

† Each value is an average of 10 specimens.

hour intervals for the next 6 hours. Daily measurements were taken after the first 8 hours. No correction was made in these values for the creep of the wood itself which would be relatively small and constant in each sample tested.

RESULTS AND DISCUSSION

Relative rigidity TBA curves for the four cured adhesives in the temperature range 25 to 200°C are shown in Figure 2. Both PRF and PPF adhesives maintain almost constant rigidity throughout this temperature range while PVA rigidity decreases substantially at about 50°C. EPI undergoes its maximum rigidity change at approximately 80°C. Apart from the TBA curves clearly differentiating between the thermo-

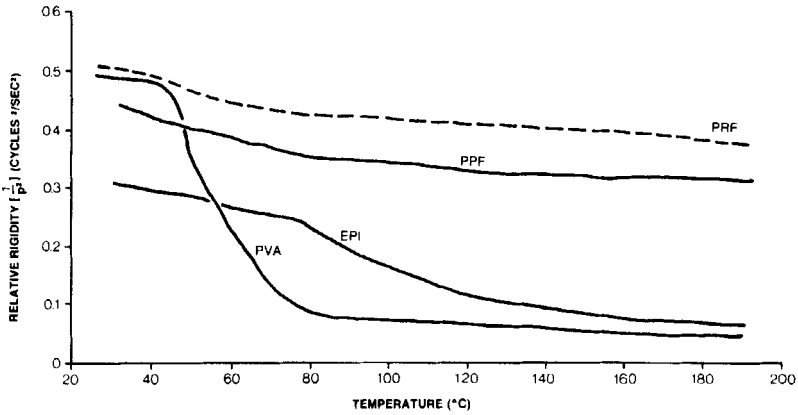

FIGURE 2 TBA rigidity curves of cured adhesives during heating at 3°C/min.

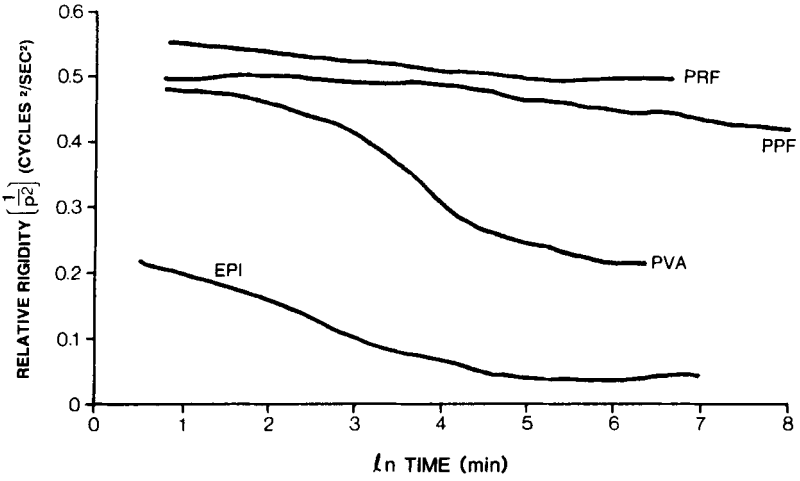

FIGURE 3 TBA rigidity curves for cured adhesives during exposure at 21°C and 92% relative humidity.

setting and thermoplastic character of these adhesives there is a difference in response of the two thermally sensitive polymers. PVA exhibits an abrupt rigidity change while the EPI initially has a much lower rigidity than the other adhesives and shows a gradual transition during heating. These PVA and EPI response differences are based in part on the molecular weight, number of crosslinks and the morphological character of these polymers[7].

Another series of TBA measurements were conducted at 21°C/92% RH. The rigidity changes observed with time (Figure 3) indicate the relative moisture sensitivity of these adhesives. At this humidity PRF and PPF show minimal change in rigidity with exposure time over a 36-hour period. During similar exposure times both PVA and EPI decreased in rigidity with PVA showing a greater change; however, its final rigidity levels were still higher than that of the EPI adhesive.

Response to thermal and moisture conditions as indicated by rigidity change demonstrates the potential for using TBA to compare adhesive properties. The data from Figures 2 and 3 clearly divide the cured resins into environment stable PRF and PPF systems and the more heat and moisture sensitive PVA and EPI systems. In addition EPI and PVA are distinguished from each other by their initial rigidity and the magnitude of the rigidity change.

TABLE II
Glueline deformation as determined by ASTM D-3535-76 test procedure

Adhesive	Total deformation (mm)	
	27°C/90% RH Exposure	71°C Exposure
PRF	<0.1	<0.1
PPF	<0.1	<0.1
EPI	1.15	2.79
PVA	failed after 50 hrs	failed after 30 hrs.

To determine the relationship between these rigidity changes and adhesive response under load, wood laminates bonded with these four adhesives were first subjected to creep tests using the ASTM D-3535-76 method. Table II gives the results of these tests which indicate that glueline deformation for PRF and PPF bonded samples was minimal. PVA samples failed at both of the high humidity and 71°C temperature exposure conditions. At both exposure conditions, EPI shows much greater creep extension than either the PRF or PPF samples but these

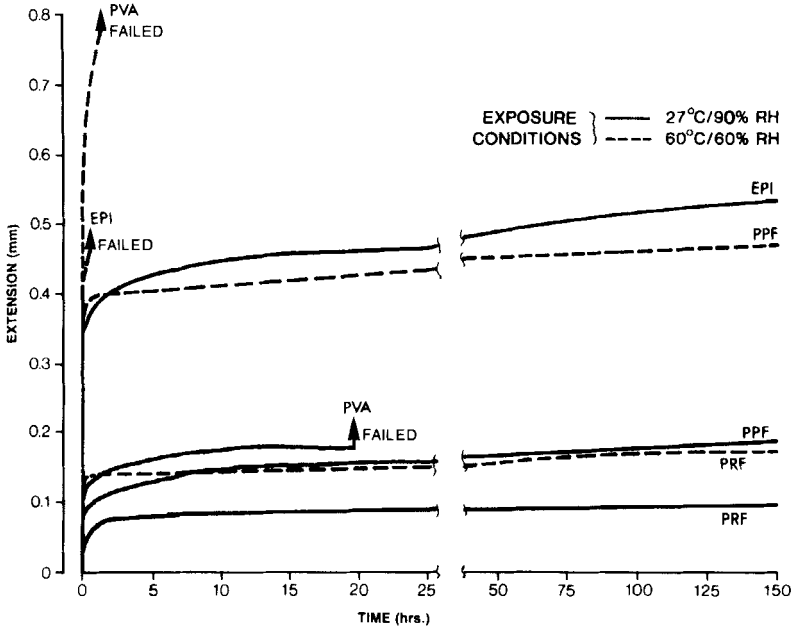

FIGURE 4 Creep extension at two exposure conditions for double shear specimens bonded with various adhesives.

values were within the 3.63 mm maximum extension allowed by the standard.

Although adhesives showing glueline deformation correspond to those with TBA rigidity and damping change, creep information in greater detail than the single extension value given by the ASTM test was needed to define this relationship better.

To accomplish this another creep experiment was conducted using double shear samples. Glueline deformations were measured over a period of 7 days and are graphically shown in Figure 4 for both 27°C/95% RH and 60°C/60% RH exposure conditions.

This latter exposure, conducted in a small kiln, was designed to determine the influence on glueline deformation of combined heat and RH. For both exposure conditions maximum rate of glueline extension occurred in the first few hours of loading. At 27°C/90% RH, PRF creeps only a minor amount, except for the initial deformation during load application. PPF shows a similar small creep extension with time while EPI exhibits the greatest extension during the testing period. Although the PVA showed only moderate creep extension, bond failure occurred after about 20 hours loading. Because of the known thermoplastic character of the PVA, this failure was expected but the relatively small glueline deformation observed at failure was somewhat surprising.

At the 60°C and 60% RH test condition, further creep response differences were noted. PRF still exhibited only a relatively minor amount of creep extension while PPF showed a greater initial extension. Both PVA and EPI adhesives failed under these loads and environmental conditions after about 1.5 and 0.5 hours loading, respectively, with the extension of the former being almost twice that of the latter. In this case, the expected thermoplastic behaviour of PVA was evident. The failure of these specimens indicates that the load level chosen was probably too high for the humidity and temperature test conditions used for these adhesives.

A qualitative relationship can be seen between the TBA data of Figures 2 and 3 and the creep results of Figure 4. For instance, PRF and PPF show minimum TBA rigidity change while also having the smaller creep extension values. EPI and PVA tend to show greatest creep extension and correspondingly exhibit large TBA rigidity changes.

Plotting the difference between the initial and final relative rigidity values from the TBA curves of each adhesive against their respective creep extension in double shear laminates after 24 hours loading time yields the trend shown in Figure 5. Here the high humidity exposure

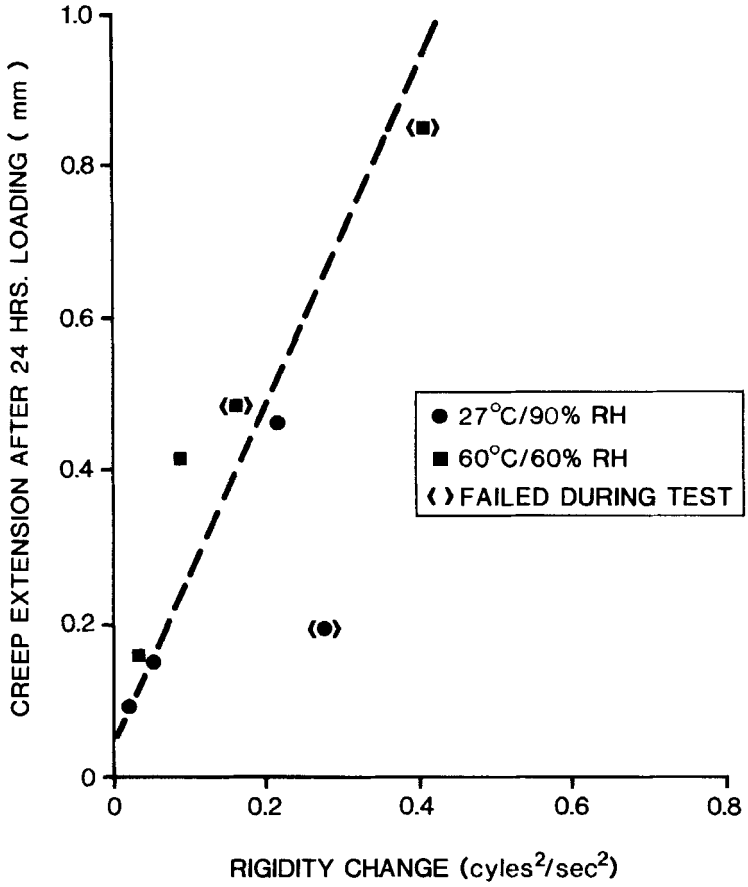

FIGURE 5 Relationship of creep extension after 24 hrs of loading to rigidity change between initial and final TBA values. Creep of failed samples are taken as maximum extension measured prior to failure.

data for both creep extension and TBA rigidity difference (Figure 3) are plotted against each other while the temperature programmed TBA rigidity difference values (Figure 2) are plotted against the 60°C/60% RH creep extension data. It is realized that comparisons between rigidity changes measured over a programmed temperature range and creep determined at one temperature may be questionable. Ideally, further experiments where creep tests are preformed at a series of different temperatures should provide more appropriate comparisons. In

this case, however, the choice of 60°C as a creep test temperature was made because this is a reasonable temperature range for accelerated aging evaluation[3] and is also near the softening temperature for the two thermoplastic type polymers. In Figure 5 creep values for the samples failing before 24 hrs are taken as the maximum extension prior to failure. One of these data points, which relates to the high humidity exposure PVA sample which failed after 20 hrs, shows only a small creep extension at large rigidity change. In contrast, the data point for the failed PVA samples exposed to 60°C/60% RH conditions shows a creep-rigidity value which follows more closely the trend of the other data points.

This observation suggests a different mechanism of failure for PVA under the two exposure conditions. Since the PVA mixture contains crosslinking agents, emulsifiers and plasticizers, these components could be influenced to varying extents by heat and moisture. At high humidity the result could be discontinuity in the PVA glueline and rapid failure without flow. At the high temperature exposure increased polymer chain motion may be the dominant effect with the resulting thermoplastic behaviour resulting in substantial creep extension prior

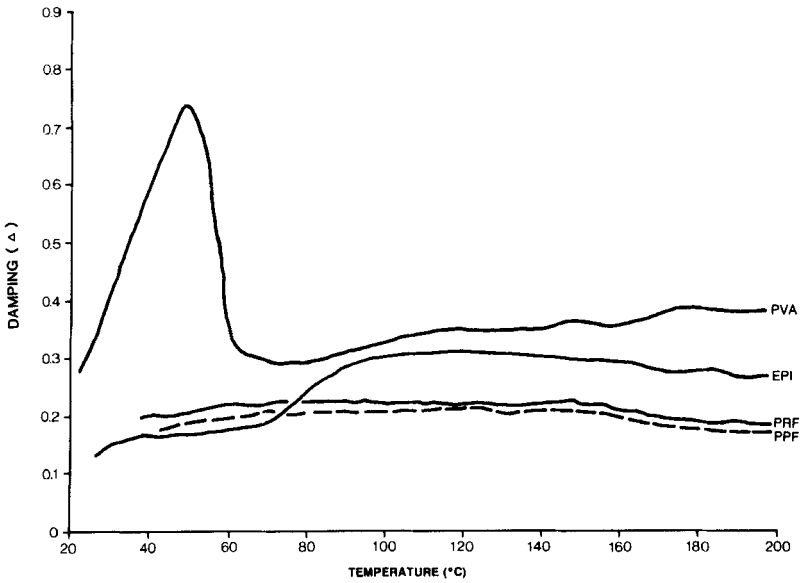

FIGURE 6 Damping loss TBA curves for cured adhesives during heating at 3°C/min.

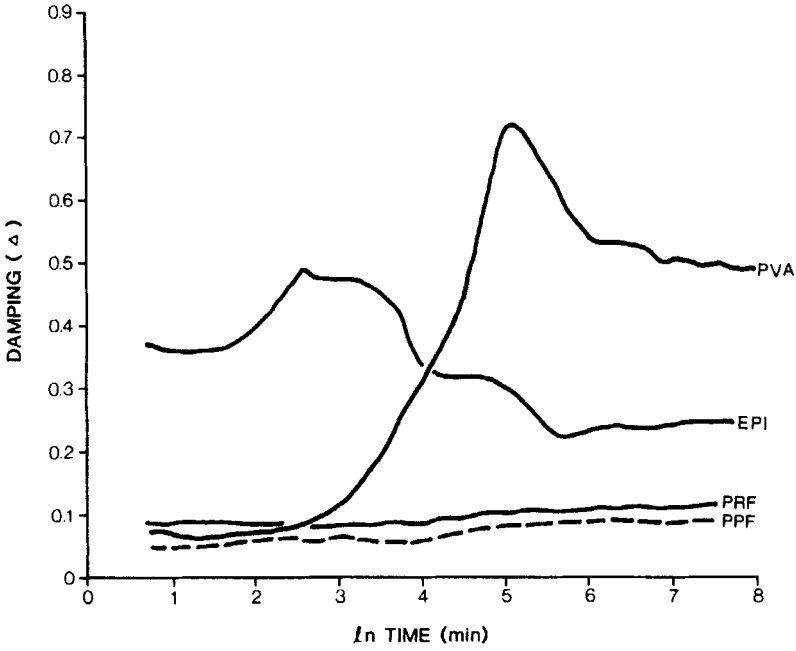

FIGURE 7 Damping loss TBA curves for cured adhesives during exposure at 21°C and 92% relative humidity.

to failure. The large rigidity change observed in all PVA samples does, however, indicate extensive physical alterations in the adhesive, which is reflected in poor performance properties such as creep or bond failure at low loading values.

Further polymer-structure information can be gained from examining the damping loss factor (Δ) portion of the TBA response during temperature and humidity runs (Figures 6 and 7). The magnitude of the damping loss factor is governed by the motion of chain segments in the polymer structure. Energy input into elastic type polymers will tend to be almost fully converted back into mechanical motion resulting in a small damping loss. In plastic (viscous flow) systems most of this energy will be dissipated as heat and will produce a large damping factor. Many thermoplastic adhesives have both a viscous and elastic component. In Figures 6 and 7, PVA shows the largest damping loss peak while EPI is next in magnitude. PRF and PPF exhibit no strong damping loss peaks. A plot of maximum damping loss value against

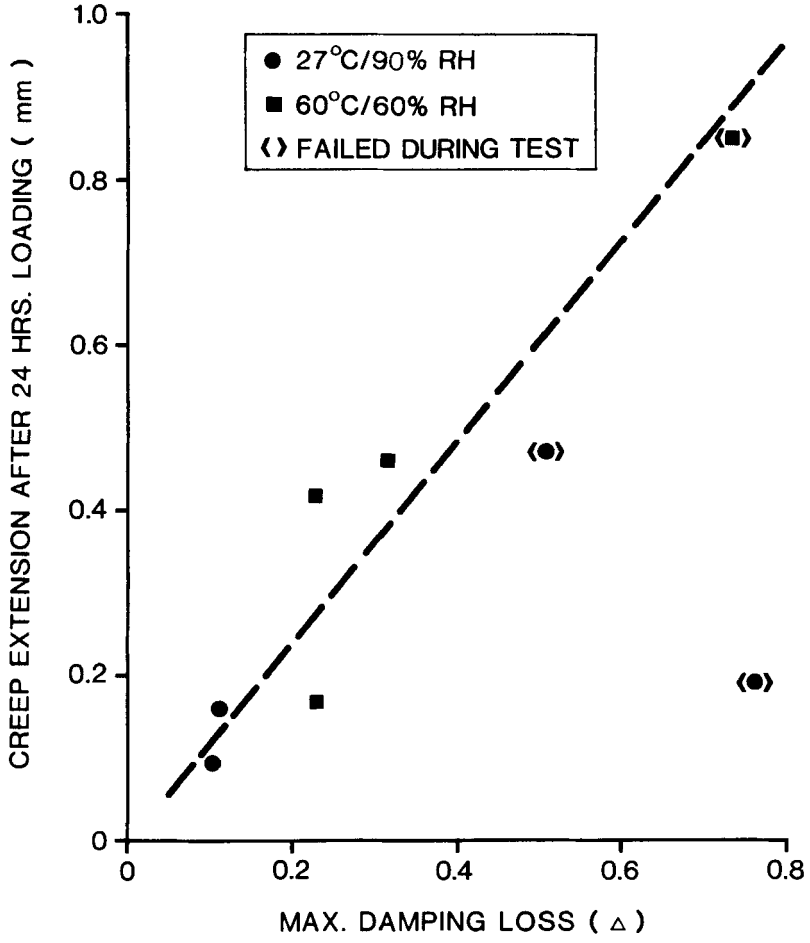

FIGURE 8 Relationship of creep extension after 24 hrs of loading to maximum TBA damping loss. Creep of failed samples are taken as maximum extension measured prior to failure.

creep extension in double shear laminates after 24 hours of loading is shown in Figure 8. Here the high humidity condition values for creep (Figure 4) and TBA damping loss data (Figure 7) are plotted against each other while the temperature programmed TBA damping loss values (Figure 6) are plotted against the 60°C/60% RH creep extension data (Figure 4). As before, creep values for samples failing before the

24 hr period are taken as the maximum extension measured prior to failure. A general positive trend again is found between the creep value and the maximum damping loss value. On the basis of large damping loss, rigidity decrease and bond failure in the creep extension test, PVA would, as experience has shown, be a poor adhesive choice for use in load bearing structures exposed to humidity and/or heat.

SUMMARY AND CONCLUSIONS

This study indicates that TBA can offer a rapid and convenient method for assessing the moisture and thermal stability of cured adhesives. Rigidity and damping loss information from the four wood adhesives considered here could, in most cases, be closely related to glueline creep behavior or bond failure in shear at high RH and moderate temperature conditions. It should be recognized that conditions of cure such as time, temperature, relative humidity and glueline thickness can alter the morphology and stability of a wood adhesive.[15] The present study only considered a few specific cure conditions. Alternate curing strategies my produce quite different polymer behavior.

In terms of evaluating the response of various adhesive types and formulations response to environmental conditions, TBA has likely applications as a prescreening method for comparing relative creep tendencies. The relative rigidity and damping loss values, however, should not be the only factor used to quantify acceptable limits for long-term adhesive performance. Performance expectations depend upon particular wood bonding service conditions and TBA provides data only on certain property characteristics. This information needs to be used together with wood bond testing to firmly establish an adhesive long-term performance capability.

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