MEASUREMENT OF UNIAXIAL CREEP OF SELECTED ADHESIVES IN FREE FILM FORM
Abstract

Satisfactory performance of adhesive-bonded wood joints results from the application of sound engineering principles and a knowledge of the mechanical behavior of the materials involved. This report describes techniques for measuring the creep of adhesives in free film form when dead loaded in tension. Creep measurements, made on selected adhesives, illustrated that adhesives differ widely in their ability to deform elastically and plastically under load. Better joint design for specific end uses can result upon considering the creep behavior of the adhesives employed.
MEASUREMENT OF UNIAXIAL CREEP
OF SELECTED ADHESIVES IN FREE FILM FORM

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Introduction

A primary requisite in the satisfactory design of an adhesive-bonded wood joint is a knowledge of the time-dependent mechanical properties of the joint and its components. For instance, an adhesive-bonded wood joint that must withstand continuous loading with a minimum of deformation for prolonged periods of time requires an adhesive that resists deformation at the prevailing stress level. The mechanical properties of this adhesive must behave in a manner that is essentially independent of time. In contrast, an adhesive whose mechanical properties are time dependent could be used for relaxing internal curing stresses or stresses caused by dimensional changes of the wood adherends. Such adhesives are capable of deforming to a relatively greater extent under stress. The time-dependent properties of the adhesives must be known before joints can be specifically designed to utilize these properties. Unfortunately, little is known about the time-dependent behavior of wood adhesives when isolated from wood joints. Meaningful data on the in-joint behavior of wood adhesives are almost as meager although Krueger2 recently developed a method of studying in-joint adhesive behavior.

1 Maintained at Madison, Wis., in cooperation with the University of Wisconsin.
The present paper presents a method for studying the time-dependent behavior of wood adhesives as free films. The work was restricted to the development of methods for measuring creep, the time-dependent deformation of a material under constant stress. Creep measurements were made on several wood adhesives at different stress levels and humidities.

In this study, adhesives were cast into thin films, and specimens cut from the films were dead loaded at various stress levels. Creep measurements were made at two relative humidities to determine the effect of moisture on time-dependent mechanical behavior. Creep of the loaded specimens was measured optically. The types of adhesives that were measured ranged from rigid, cross-linked materials to soft viscoelastic materials. The majority of creep measurements were confined to the more deformable viscoelastic types of adhesive, since the optical measuring system lacked sufficient sensitivity to measure the minute creep of the rigid, cross-linked adhesives with accuracy.

The methods used in casting films of adhesives and testing the films in creep were not unique to this study. The time-dependent mechanical properties of many other kinds of polymeric materials are frequently determined from free-film specimens using dead loads. Creep measurements on film specimens are commonly made electrically with displacement transducers and strain gages, or visually observed with microscopes and telescopes. A visual measuring system using a cathetometer was employed in this study, because a high degree of sensitivity and precision was not mandatory for measuring the large deformations characteristic of the majority of adhesives being tested.

Data obtained in this study have been analyzed to point out the major differences in creep behavior of wood adhesives under different stress levels and humidities. No sophisticated mechanical analysis of the creep data has been attempted because of the somewhat exploratory nature of the study. However, the literature is extensive concerning the theories, mathematics, and data interpretation of creep behavior of polymeric materials in terms of both phenomenological and molecular aspects. Much of this information is applicable to wood adhesives. Literature on the mechanical properties of polymeric materials is thoroughly reviewed by Alfrey, Tobolsky, Baer, and Thorkildsen. Thorkildsen’s work also includes an extensive bibliography on creep studies of plastic materials.

Procedure

**Adhesive Components**

The material components used in the study were adhesives, extenders, and hardeners. The various formulations of adhesives are listed in table 1.

Both of the polyvinyl acetate (PVA) emulsions were commercial adhesives. The unmodified PVA adhesive was a water emulsion with approximately 50 percent solids content, and the thermosetting PVA adhesive was a water emulsion containing 48 percent solids. The latter adhesive required a catalyst and developed a certain degree of cross-linking upon aging or after heating.

The resorcinol adhesive was a commercial phenol-resorcinol resin solution to which was added a commercial hardener consisting of approximately 50 percent paraformaldehyde and 50 percent walnut shell flour (by weight).

The epoxy resin was a commercial liquid adhesive with an epoxide equivalent of 185–192. Epoxide equivalent is the number of grams of resin containing 1 gram equivalent of epoxide.

Both the polysulfide extender and the polyamide extender were commercially available products. The polyamide was of the branched chain, reactive type, and was very viscous.

**Preparation of Specimens**

The geometry of the specimen (fig. 1) used for creep measurements was a rectangular strip approximately 5 inches long, 0.25 inch wide, and uniform in thickness. Films ranging in thickness from 0.005 to 0.028 inch were cast depending on the viscosity of the adhesive. V-shaped gage marks were drawn with India ink 0.5 inch apart near the center of the specimen to provide reference points for measuring deformations. Before making a test the marked portion of the specimen was centered between the clamps, which were separated by a fixed initial spacing of 2 inches. Measuring creep at the center of the specimen and over such a short gage length had a distinct advantage. The stress distribution was known to be more uniform near the center of the specimen than near the clamps. Also, the neck-down due to straining was essentially uniform in the center area of the specimen.

Each of the five adhesive types required slightly different film-casting and specimen-cutting techniques. The details of the specimen-preparation techniques used in each case were as follows:
Polyvinyl-acetate-emulsion.--A sheet of clean plate glass was waxed with a general-purpose solvent-type paste wax. Care was taken to spread the wax uniformly and smoothly.

The adhesive was thoroughly stirred at room temperature. Walnut shell flour, when used as a filler, was added at this point. The adhesive was then poured onto the waxed glass plate and spread into a film using a 6-inch adjustable doctor blade (Boston-Bradley adjustable blade; Gardner Laboratories, Bethesda, Md.\(^2\)) set at 0.020 inch (fig. 2). It was essential that the doctor blade be pulled at a uniform rate, otherwise the resulting film would vary in thickness.

The film was allowed to dry slowly in an 80° F., 80 percent relative humidity environment until it reached a soft, leathery state. Specimens could be cut readily from films in this state, but not from brittle, completely dried films. Specimens, 1/4 inch wide, were cut from the partially cured film using a razor blade and a metal guide as shown in figure 3. By running the razor blade through a slot in the guide, both edges of the film were supported during cutting. If only one edge was supported, checks frequently developed in the unrestrained edge.

Thermosetting polyvinyl-acetate emulsion.--The procedure for casting thermosetting polyvinyl-acetate differed from that used with unmodified polyvinyl acetate in two ways. The thermosetting adhesive required the addition of a hardener and dilution by about 20 percent with water. After the formulation was thoroughly mixed it was allowed to stand for an hour before casting. The dilution reduced the viscosity of the formulation so that the air bubbles entrained by stirring could combine and rise to the surface. Dilution, followed by gentle centrifuging, was equally successful in reducing the size and number of bubbles in the dried films to an acceptable level.

Phenol-resorcinol resin.--Two glass plates were waxed, and 0.012-inch aluminum shims were placed around the edges of one plate. After thoroughly blending the resin with its hardener, the mixture was poured along one side of the shimmled glass plate. The second plate was slowly lowered from one side (scissor-type action) so that the adhesive was squeezed out toward the center of the plates (fig. 4). When carefully done, few air bubbles were trapped in the film, because the advancing adhesive forced out the air between the plates. Weights were placed on the plates, and the film was allowed to cure overnight. The partially cured film was cut into specimens in the manner described for polyvinyl acetate.

\(^2\)The mention of firm names and trade products does not imply that they are endorsed or recommended by the Department of Agriculture over other firms or similar products not mentioned.
Phenol-resorcinol films could not be prepared on a single plate, as were the polyvinyl acetates, because the film warped during the drying and curing stages. Internal stresses resulting from unequal drying and curing rates caused the rigid phenol resorcinol to warp permanently, whereas the viscoelastic nature of the polyvinyl acetates allowed stress relaxation to occur and prevented permanent warping.

**Epoxy extended with polysulfide.**—Two glass plates were coated with a silicone mold-release agent, and aluminum shims 0.013 inch thick were placed around the edges of one plate. The epoxy resin and polysulfide extender were mixed together in the desired proportions and then heated to approximately 212° F. The heating served two purposes: (1) It reduced the viscosity of the mixture enough to allow air bubbles to combine and rise to the surface; and (2) the reduced viscosity facilitated spreading of the adhesive into a film. After most of the bubbles disappeared, the catalyst was stirred in and the hot adhesive was immediately poured along one side of the shimmed glass plate. The top plate was lowered onto the adhesive using the scissor-type action described for casting phenol resorcinol. Heavy weights were placed on the glass, and the film was allowed to cure completely before removal.

The films were cut into specimens using a sharp papercutter. However, the films containing 80 parts and 60 parts of epoxy resin were slightly brittle and had to be softened in a stream of hot air from an electric dryer (heat gun) while being cut.

**Epoxy extended with polyamide.**—The procedure for casting polyamide-extended epoxy was identical to that used for polysulfide-extended epoxy except the materials required heating separately before mixing, because of the reactive nature of these components when combined. After combining the two components, the hot mixture was allowed to stand for several minutes to give air bubbles an opportunity to rise to the surface.

**Specimen Conditioning and Aging**

All specimens were stored in an 80° F., 30 percent relative humidity environment until tested. The polyvinyl-acetate specimens required special treatment in that they had to be kept flat between two glass plates for a week after casting. Thermosetting polyvinyl acetate required over a month to approach complete cure, whereas unmodified polyvinyl acetate and the other adhesive types appeared to be adequately cured after a week or less. Specimens tested at 65 percent relative humidity were conditioned to that humidity for several days before being used. Whenever possible, specimens of a particular adhesive were tested after conditioning and aging for approximately equal periods.
Specimen Examination, Marking, and Measurement

All specimens were carefully examined under a stereomicroscope (40X) and those having checks, large or excessive air bubbles, or other serious discontinuities were discarded. Strict selection for homogeneity was very important in reducing creep variations caused by physical flaws.

The V-shaped gage marks were drawn on each specimen using a fine-pointed pen and india ink. India ink proved to be a desirable marking agent because of its opacity, adhesion, and ability to deform along with the creeping adhesive.

A micrometer was used to measure width and thickness between the gage marks to the nearest 0.001 inch. Cross-sectional area was computed to the nearest 0.0001 square inch using the averages of three separate width and thickness measurements. Distance between gage marks was measured with a cathetometer (10X) to the nearest 0.01 centimeter, or in some cases with a traveling microscope to the nearest 0.001 centimeter.

Creep Measuring Apparatus

The steel grips fabricated to hold film specimens during creep tests are shown in figure 5. The lower grip slides freely on guide rods attached to the upper grip to minimize unwanted lateral and torsional forces acting on the specimen. Small pins were inserted into holes bored in the ends of the guide rods to prevent the lower grips from falling off and striking neighboring grips when the specimens failed. Fine-grit aluminum-oxide abrasive cloth glued to the inside faces of the grips helped to prevent specimens from slipping under heavy loads. An aluminum gage block (fig. 5) was used to center the specimens in the grips and to insure a constant initial spacing of 2 inches between the grips.

Two racks were constructed to support the grips—one in an 80° F., 30 percent relative humidity environment, the other in an 80° F., 65 percent relative humidity environment. Each rack consisted of a rigid steel beam fitted with 20 hooks spaced 6 inches apart to provide positions for the grips to hang. The effects of vibrations coming from the floor and walls were reduced by placing foam rubber under the beams. A shelf below the beams supported the specimen-loading device and served as a platform for the cans of lead shot (loads) to fall upon when specimens failed (fig. 6).

An electric time totalizer was connected with each grip on the racks via a mechanical-electrical system. A lever-actuated microswitch was mounted above
each position on the rack, as shown in figure 7. Nylon monofilament was fastened to the microswitch lever arm, passed through a screw eye, and was tied to the pin of the lower grip. When a specimen failed, the weight of the lower grip pulled the line taut, actuating the microswitch and stopping the time totalizer.

Creep measurements were made with a cathetometer as shown in figure 8. (An earlier version of the mechanical–electrical system is also shown). The cathetometer was moved to within 6 feet of the specimens and the positions of the specimen gage marks were read from the calibrations on the support standard to the nearest 0.01 centimeter. A traveling microscope (accurate to 0.001 in.) was used in some instances when very little creep occurred. An important limitation of the visual method of measuring creep was the finite amount of time that elapsed between reading the upper and lower gage marks. During this time interval, the specimen continued to creep, with the initial positions of the gage marks changing accordingly. However, the error thus incurred was not serious in the majority of creep tests because of the low rates of creep involved.

Specimens were dead–loaded to the desired stress levels (ranging from 150 to 3,000 pounds per square inch) with cans containing lead shot. When specimens were tested at high stress levels, the loads were applied slowly with the aid of a screw-type adjustable platform (fig. 6). A slow and uniform rate of loading reduced the number of failures that occurred during loading and improved reproducibility of initial creep data.

Calculations

Creep was calculated as the change in deformation between the gage marks as a function of time. However, for comparison purposes creep strain was found to be a more useful quantity. Creep strain was calculated as follows:

$$\varepsilon_t = \ln \frac{L}{L_0}$$

where \(\varepsilon_t\) is true strain, \(L_0\) is initial gage length before loading, and \(L\) is gage length at any time \(t\) after loading. The value of \(\varepsilon\) in this equation is called “true strain” since it is based on a changing gage length rather than on a constant length.8 True strain was used because of the large deformations that frequently occurred with the stress levels and materials that were used.

All of the creep data recorded in the study was plotted as creep curves showing creep strain versus time. The slopes of the curves indicated the rate of creep. Comparisons among the various adhesives were made on the basis of the initial slope values of their creep curves (fig. 9). Initial slope, or creep rate, was chosen as the basis of comparison for two reasons. First, it was a value that could be obtained from the low-strain portion of the creep curve where the stress level had not been excessively increased by the reduction in cross-sectional area that accompanies strain. Secondly, initial creep rate was the only rate value that could be obtained in a reasonable length of time for materials that crept very slowly. Unfortunately, the initial portions of some creep curves were far from linear, which resulted in slope values that were somewhat arbitrary. Initial creep rates for the majority of data were computed as fitted regression lines using the least-squares method.

The actual or true stress acting on a creeping film specimen increased as the specimen elongated and its cross-sectional area decreased. True stress was calculated using the following equation:

\[
\sigma_t = \frac{l}{L_0} \sigma
\]

where \( \sigma_t \) is true stress, and \( \sigma \) is nominal stress. This equation assumes that the volume of material under stress remains constant.

Results

Polyvinyl Acetate Adhesives

Typical creep curves for polyvinyl acetate at stress levels of 250, 500, 750, and 1,000 pounds per square inch are shown in figure 10. Creep curves of the other polyvinyl acetate formulations were essentially similar in shape. These curves were obtained with a constant applied load. The stress level, or load per unit cross-sectional area, increased as the specimens elongated and shrank in cross-sectional area. The true stress at the point of maximum strain is also shown for each curve.

\footnote{Large strains were considered to be greater than 50 percent nominal strain in this study. Theoretically, the stress level should be kept constant for creep tests by decreasing the load in accordance with the amount of strain; but because of the practical difficulties involved, the stress is frequently just assumed to be constant. However, in this study it was considered unrealistic to ignore the change in stress when nominal strains exceeded 50 percent.}
A summary of creep rate data obtained from all the polyvinyl acetate formulations is presented in table 2. Initial creep rates are listed for each stress level and relative humidity. Both the ranges and average values of the creep rate data are presented. The factor of increase in creep rate resulting from exposure to 65 percent relative humidity as compared with 30 percent relative humidity is shown for each adhesive at each stress level.

Figures 11 and 12 show the effect of stress level on the creep rates of the extended and unextended polyvinyl acetates at conditions of 80° F., 30 percent relative humidity, and 80° F., 65 percent relative humidity, respectively.

Creep curves of thermosetting PVA aged 9, 31, and 57 days before testing at a constant nominal stress level of 2,500 pounds per square inch are shown in figure 13. The initial creep rate is written beside each curve.

Figure 14 shows how the creep behavior of thermosetting PVA was affected by curing the film at 150° F. for various periods of time up to 6 hours before testing. Total creep strain after 2 hours is plotted against the length of time the film was cured at 150° F.

A typical creep and recovery curve for unmodified polyvinyl acetate at 1,000 pounds per square inch and 30 percent relative humidity is illustrated in figure 15. The regions of elastic and viscoelastic behavior are labeled.

**Phenol–Resorcinol and Epoxy Adhesives**

Typical creep curves for the polysulfide-extended epoxy formulations measured at 65 percent relative humidity are plotted in figure 16; similar curves for polyamide-extended epoxy resin measured at 30 percent relative humidity are given in figure 17. Stress levels of 150, 1,000, and 1,500 pounds per square inch were used in both adhesive films for extensions of 50, 40, and 30 percent respectively.

Table 3 is a summary of creep rate data obtained from the phenol–resorcinol and polysulfide-extended epoxy adhesives. Initial creep rates are listed for each stress level and relative humidity. The factor of increase in creep rate resulting from exposure to 65 percent relative humidity as compared with 30 percent relative humidity is shown for each adhesive.

Table 4 is a summary of creep strain data for polyamide-extended epoxy. The amount of strain that occurred after 120 hours is shown for each formulation at relative humidities of 30 and 65 percent and stress levels of 150, 1,000, and 1,500 pounds per square inch.
Discussion

Polyvinyl Acetate Adhesives

The shape of the creep curves for the polyvinyl acetate adhesives under different stress levels (fig. 10) is typical for plastic materials. These curves begin with an essentially instantaneous deformation that occurs upon application of the load. This deformation is composed of initial elastic and plastic strains, which are completely recoverable upon removal of the load, and whose magnitude is proportional to the stress level. The second region of the curves is characterized by an increasing slope (creep rate) at high stress levels and a decreasing slope at low stress levels. The third region, when present, is characterized by a relatively constant rate of creep. This region of constant creep rate is reached most rapidly at high stress levels as shown in figure 10. A fourth region of decreasing creep rate is evident at the high stress levels, which indicates that an internal stiffening of the material occurs at large deformations.

The initial creep rates of the different polyvinyl acetate formulations differed considerably depending on the stress level and humidity (table 2). For instance, unmodified PVA at 30 percent relative humidity crept 12 times faster at a stress of 1,000 pounds per square inch than at 250 pounds per square inch, whereas thermosetting PVA crept only 4 times faster at 1,000 p.s.i. than at 250 p.s.i. (fig. 11). This difference indicates that the thermosetting PVA at 30 percent relative humidity is several times more resistant to creep at these stress levels than is the unmodified PVA. Similar creep rate comparisons made at 65 percent relative humidity show that unmodified PVA crept only 6 times faster at 1,000 p.s.i. than at 250 p.s.i., whereas thermosetting PVA crept over 1,000 times faster at 1,000 p.s.i. than at 250 p.s.i. It appears that at 65 percent relative humidity the unmodified PVA has been so plasticized by atmospheric moisture that it creeps very rapidly even at the low stress of 250 p.s.i. Thermosetting PVA, on the other hand, appears to be able to resist creep at 250 p.s.i. and 65 percent relative humidity quite well. But, when the stress level was increased to 500 p.s.i., the creep rate increased 100 times over its value at 250 p.s.i. This large increase in creep rate, which was caused by a relatively small increase in stress level, suggests the existence of a limiting stress level below which the material can satisfactorily resist creep at 65 percent relative humidity. The limiting stress level for thermosetting PVA at 65 percent relative humidity must lie between 250 and 500 p.s.i., whereas unmodified PVA at 65 percent relative humidity must have a limiting stress level considerably below 250 p.s.i. The creep rate of thermosetting PVA begins to approach that of unmodified PVA once the limiting stress level has been exceeded (fig, 12).
At 30 percent relative humidity, the thermosetting PVA appears to be below its limiting stress level even at 1,000 p.s.i., whereas unmodified PVA at 1,000 p.s.i. may already have reached its limiting stress level (fig. 11).

Table 2 shows the factor of increase in creep rate from 30 percent to 65 percent relative humidity for each adhesive at each stress level. Note that thermosetting PVA at 250 p.s.i. was the only combination of adhesive and stress level, that could withstand the increase in humidity. In contrast, thermosetting PVA at 1,000 p.s.i. exhibited the greatest change in creep behavior with the increase in humidity.

The effect of adding 10 percent walnut shell flour to the PVA adhesives is shown in figures 11 and 12. At 30 percent relative humidity and 250 p.s.i., the extended adhesives crept less rapidly than the unextended adhesives, but at 500 and 1,000 p.s.i. the reverse was true. The difference in creep rates at 250 p.s.i. is difficult to explain, but at the higher stress levels the difference may be the result of having less adhesive per unit volume of film. The walnut shell flour acts as an impurity in the film which reduces the effective cross-sectional area that must support the load. A reduced cross section results in an increased stress level and creep rate. However, the same explanation is not valid for the extended PVA adhesive at 65 percent relative humidity, since the extender reduced the creep rate. The influence of walnut shell flour on creep behavior at 65 percent relative humidity may have been to interfere with the uniform plastic flow of the PVA resin systems. If such interference were great enough, it could have reduced the rate of creep.

The superior creep properties of thermosetting PVA resulted from chemical crosslinks that formed between the polymer molecules upon aging and/or heating. These crosslinks helped to resist the gradual slipping of molecules past one another when stress was applied. In addition, the crosslinks between polymer molecules reduced the plasticizing effect of adsorbed moisture. Unmodified PVA was not a cross-linked material, and therefore exhibited more rapid creep and less moisture resistance than thermosetting PVA.

The creep behavior of thermosetting PVA was highly dependent on the age of the film at the time of test. As the film grows older initial creep rate decreases and time to failure increases (fig. 13); the films showing initial creep rates after 9, 31, and 57 days' aging of 0.0027, 0.013, and 0.002 inches per inch per hour respectively. The cross-linking reaction in thermosetting PVA is very slow.

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10The equilibrium moisture content of wood is 6 percent at 80° F., 30 percent relative humidity, and 12 percent at 80° F., 65 percent relative humidity. In many parts of the United States, glued wood constructions used in interior applications may vary in moisture content from 6 to 12 percent throughout the year.
Creep behavior is a very sensitive indicator of the degree of cure, and figure 14 illustrates how creep behavior was used to determine the cure of thermosetting PVA exposed to 150°F for varying periods of time. After 6 hours at 150°F, the adhesive had not reached full cure, even though the initial creep rate already equaled that of 78-day-old film cured at room temperature. This experiment of heat curing versus room-temperature curing of thermosetting PVA suggests that the degree of cure that can be obtained using high temperatures may exceed that which is possible at room temperatures. Further investigation will be required to confirm this suspicion. The creep behavior of unmodified PVA indicated that its mechanical properties reached a steady state soon after film formation, and the films could be used in creep tests as soon as a week after casting.

Both thermosetting and unmodified PVA exhibited variability in their creep behavior between batches and between specimens from the same batch. Thermosetting PVA was far more variable than unmodified PVA, even for specimens of the same size and origin. However, as the thermosetting PVA specimens grew older, their variability appeared to decrease, which suggests that some of the specimens may have cured more rapidly than others. Unequal curing of the film could have resulted from a nonuniform distribution of hardener, although every batch was thoroughly mixed before film casting.

Typical recovery behavior of the polyvinyl acetates is shown in figure 15. Even at relatively small strains, these materials were characterized by large amounts of permanent deformation; that is, they did not recover to their original dimensions after the load was removed. Also, the amount of recoverable strain appeared to decrease as humidity increased, indicating that the PVA materials were being plasticized by the atmospheric moisture.

**Epoxy and Phenol-Resorcinol Adhesives**

Creep studies made on epoxy resin extended with polysulfide and polyamide additives were more exploratory in nature than the studies conducted on the polyvinyl acetate adhesives. Each epoxy formulation was tested at only one stress level instead of several. The magnitude of the stress used for each formulation was selected by trial and error with the purpose of producing a measurable amount of creep within a reasonable length of time.

Reproducible creep behavior was very difficult to obtain with the extended epoxy films. One reason for inconsistent creep behavior may have been non-uniform composition of the films, since the resin and extenders were viscous.
and difficult to mix. Another reason may have been variable cure in the films resulting from differences in heat treatment during casting and curing. Creep behavior also varied with age, because the films continued to cure at room temperatures.

Epoxy-polysulfide films were much more consistent in creep behavior than the epoxy-polyamide films. The reason for this difference may have been a more uniform composition of the epoxy polysulfide, since the polysulfide was less viscous than the polyamide and could be thoroughly mixed with the epoxy. Typical creep curves for the epoxy-polysulfide formulations are shown in figure 16. Note that the 50–50 formulation at only 150 p.s.i. exhibited a large initial elastic strain, rapid creep, and early failure, whereas the 80–20 formulation at 1,500 p.s.i. exhibited practically no initial elastic strain and no creep over the same time period. The 60–40 formulation crept less and lasted longer than the 50–50 formulation even though it was under a stress level almost seven times greater. The initial creep rates of the epoxy-polysulfide formulations increased when the relative humidity was raised as shown in table 3. The 60–40 formulation appeared to be the most sensitive to humidity,

Creep rate values for phenol-resorcinol adhesive at 3,000 p.s.i. are also shown in table 3. The creep rate of the phenol-resorcinol was extremely slow even though the adhesive was under a stress so great that almost all other adhesive formulations would have failed immediately.

The creep rate of the phenol-resorcinol appears to increase greatly as the relative humidity increases from 30 percent to 65 percent, although it is still relatively small in magnitude at 65 percent relative humidity.

The creep behavior of the epoxy-polyamide formulations varied greatly even for specimens from the same film. Such variable behavior probably resulted from a nonuniform distribution of the polyamide throughout the film. The polyamide was extremely viscous and could not be mixed with the epoxy resin (also viscous) unless both components were hot. In addition, the epoxy resin and polyamide quickly react with each other even without a catalyst. Examples of creep curves for the different epoxy-polyamide formulations are shown in figure 17. The 50–50 polyamide formulation at 150 p.s.i. did not exhibit the large initial elastic strain and early failure that was characteristic of the 50–50 polysulfide formulation. The 60–40 polyamide formulation showed almost the same initial elastic strain as its polysulfide counterpart, but it neither crept as rapidly nor failed as soon. Both of the 80–20 formulations exhibited little, if any, creep at 1,500 p.s.i. The effect of humidity on the creep behavior of the epoxy-polyamide formulations is shown in table 4. The amount of creep strain reached after 120 hours was used for comparison purposes, because the
extremely variable creep behavior of the films made it impossible to obtain meaningful initial creep rate values. All of the polyamide formulations crept more at 65 percent relative humidity than at 30 percent relative humidity, but the 60–40 formulation showed the greatest increase.

Exploratory recovery measurements made on the 50–50 and 60–40 epoxy-polysulfide formulations showed total recoveries of 95 percent and 80 percent, respectively, after a creep strain of 30 percent had been reached. Corresponding formulations of epoxy-polyamide showed total recoveries of only 65 percent and 35 percent, respectively, after a creep strain of 16 percent had been reached. This demonstrates the fact that the polysulfide is the more elastomeric of the two extenders.

Conclusions

The creep behavior of many adhesive materials can be easily studied using the apparatus and methods described in this report. The optical system employed for measuring creep worked well at moderate creep rates, but was not practical for extremely slow or fast rates of creep.

On the basis of creep data obtained on polyvinyl acetate adhesives, it was concluded that unmodified PVA crept more rapidly than did the fully cured thermosetting PVA at the stress levels and humidities used in this study. Both types of PVA adhesive were plasticized by moisture, and therefore exhibited large increases in creep rates at high humidities. Adding walnut shell flour as an extender to the adhesives increased their creep rates at 30 percent relative humidity but decreased their creep rates at 65 percent relative humidity. The superior creep resistance of the fully cured thermosetting PVA was attributed to its cross-linked structure. Creep behavior was a very sensitive indicator of the amount of cure or crosslinking that developed in the film as it aged.

Extending epoxy resin up to 50 percent with the polysulfide additive increased its flexibility, permitted large creep strains that were almost completely recoverable, and reduced its strength. Extending epoxy resin up to 50 percent with the polyamide additive produced the same general effects as extending with the polysulfide additive except less recovery was obtained after large creep strains. Both types of extended epoxy increased in creep rate with an increase in humidity, but nowhere near the extent exhibited by the PVA adhesives. Data from the exploratory creep studies on extended epoxy adhesive indicated the need for additional creep information using different stress levels and different amounts and kinds of elastomeric extenders.

The rate of creep of the phenol-resorcinol adhesive was practically negligible compared to that found for the other adhesives.
Table I. Summary of adhesive formulations used for film casting

<table>
<thead>
<tr>
<th>Resin</th>
<th>Extender</th>
<th>Hardener</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
<td>Parts</td>
<td>Type</td>
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<tr>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>PVA emulsion</td>
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<td>None</td>
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<tr>
<td>Thermostatic PVA emulsion</td>
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<td>None</td>
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<td>Resorcinol resin solution</td>
<td>90</td>
<td>Walnut shell</td>
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<tr>
<td>Epoxy resin</td>
<td>80</td>
<td>Polysulfide</td>
</tr>
</tbody>
</table>

1. Weight includes the volatile portions of the emulsions and solutions.
2. Polyvinyl acetate.
3. Diethylenetriamine.
Table 2.--Summary of creep-rate data for unmodified and thermosetting polyvinyl-acetate adhesives

<table>
<thead>
<tr>
<th>Adhesive</th>
<th>Stress level</th>
<th>Initial creep rate at 80° F., 30 percent RH</th>
<th>Initial creep rate at 80° F., 65 percent RH</th>
<th>Factor of increase</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>P.s.i.</td>
<td>In. per in. per hr.</td>
<td>In. per in. per hr.</td>
<td></td>
</tr>
<tr>
<td>Unmodified PVA</td>
<td>250</td>
<td>$2\times0.23 -0.44$ (7)</td>
<td>$2\times0.756-1.030$ (8)</td>
<td>2,580</td>
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<tr>
<td></td>
<td>500</td>
<td>$.60 - .86$ (5)</td>
<td>3,010-4,070 (8)</td>
<td>4,800</td>
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<tr>
<td></td>
<td>1,000</td>
<td>2.99 -5.66$ (8)</td>
<td>4,800-6,000 (4)</td>
<td>1,250</td>
</tr>
<tr>
<td>Thermosetting PVA</td>
<td>250</td>
<td>.001- .028 (5)</td>
<td>.39- .50 (5)</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>.030- .060 (5)</td>
<td>39.2 -57.7 (4)</td>
<td>835</td>
</tr>
<tr>
<td></td>
<td>1,000</td>
<td>.090- .090 (5)</td>
<td>373-672 (6)</td>
<td>528</td>
</tr>
<tr>
<td>Extended PVA</td>
<td>250</td>
<td>.71 - .89 (3)</td>
<td>386-461 (8)</td>
<td>500</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>1.39 -2.26 (5)</td>
<td>1,500-3,630 (8)</td>
<td>1,210</td>
</tr>
<tr>
<td>Extended thermosetting PVA</td>
<td>250</td>
<td>0 - .01 (4)</td>
<td>.88- 1.39 (6)</td>
<td>107</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>.039- .074 (5)</td>
<td>23.3 -32.3 (6)</td>
<td>443</td>
</tr>
<tr>
<td></td>
<td>1,000</td>
<td>.390 - .696 (5)</td>
<td>340-553 (6)</td>
<td>912</td>
</tr>
</tbody>
</table>

1The number of times by which the creep rate at 30 percent RH must be multiplied to obtain the creep rate at 65 percent RH.

2Values in parentheses indicate number of specimens included in the range.

3Extended 10 percent by weight with walnut shell flour.

4Estimated creep rate. Amount of creep that occurred was below the sensitivity of the optical measuring system.
Table 3.--Summary of creep-rate data for phenol-resorcinol and epoxy-polysulfide adhesives

<table>
<thead>
<tr>
<th>Adhesive</th>
<th>Stress level</th>
<th>Initial creep rate at 80° F., 30 percent RH</th>
<th>Initial creep rate at 80° F., 65 percent RH</th>
<th>Factor of increase</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>P.s.i.</td>
<td>In. per in. per hr.</td>
<td>In. per in. per hr.</td>
<td></td>
</tr>
<tr>
<td>Phenol-resorcinol</td>
<td>3,000</td>
<td>2.000177-</td>
<td>0.00180</td>
<td>0.105-0.223(2)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>.00183(2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>80 percent epoxy - 20 percent polysulfide</td>
<td>1,500</td>
<td>--</td>
<td>negligible³</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>60 percent epoxy - 40 percent polysulfide</td>
<td>1,000</td>
<td>11.6-17.2(5)</td>
<td>14.0</td>
<td>116-145(4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>50 percent epoxy - 50 percent polysulfide</td>
<td>150</td>
<td>32.3-46.9(5)</td>
<td>41.1</td>
<td>92-153.0(5)</td>
</tr>
</tbody>
</table>

¹The number of times by which the creep rate at 30 percent RH must be multiplied to obtain the creep rate at 65 percent RH.

²Values in parentheses indicate number of specimens included in the range.

³Amount of creep that occurred, if any, was below the sensitivity of the optical measuring system.
Table 4.--Summary of creep-strain data for epoxy-polyamide adhesives

| Adhesive             | Stress level | Strain reached after 120 hours at 30 percent RH | Range | Average | Strain reached after 120 hours at 65 percent RH | Range | Average | Factor of increase
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>80 percent epoxy -</td>
<td>1,500</td>
<td>0.010 - 0.020(6)</td>
<td>0.013</td>
<td></td>
<td>0.010 - 0.040(6)</td>
<td>0.020</td>
<td></td>
<td>1.5</td>
</tr>
<tr>
<td>20 percent polyamide</td>
<td>1,000</td>
<td>.005 - .010(6)</td>
<td>.009</td>
<td></td>
<td>.086 - .190(5)</td>
<td>.145</td>
<td></td>
<td>16.1</td>
</tr>
<tr>
<td>40 percent epoxy -</td>
<td>1,000</td>
<td>.010 - .010(5)</td>
<td>.014</td>
<td></td>
<td>.020 - .148(6)</td>
<td>.071</td>
<td></td>
<td>5.1</td>
</tr>
<tr>
<td>50 percent epoxy -</td>
<td>150</td>
<td>.010 - .020(5)</td>
<td>.014</td>
<td></td>
<td>.020 - .148(6)</td>
<td>.071</td>
<td></td>
<td>5.1</td>
</tr>
</tbody>
</table>

1. The number of times by which the strain at 30 percent RH must be multiplied to obtain the strain at 65 percent RH.
2. Values in parentheses indicate number of specimens included in the range.
Figure 1.—Specimen of free film adhesive for creep measurement.
Figure 2.—An adjustable doctor blade was used to spread films of polyvinyl acetate on waxed glass plates.
Figure 3.--Polyvinyl acetate film being cut into creep specimens. Cutting the film through the slot in the guide reduced checking in the edges of the specimens by supporting both sides of film while it was being cut.
Figure 4.--Phenol-resorcinol and epoxy adhesive films were formed by a scissor-type action between two waxed glass plates separated by shims.
Figure 5.--Two sets of specimen grips and aluminum gage block. The lower grip assembly shows an untested polyvinyl acetate specimen; the upper grip assembly shows a similar specimen that has crept 300 percent.
Figure 6.--Apparatus for supporting and loading film specimens; also shown are screw-type load applicator, bank of time totalizers, and electrical microswitches mounted above the specimen grips.
Figure 7.--Mechanical-electrical switching system used to stop electrical time totalizers when specimen failed.
Figure 9. -- Method of determining initial creep rate. Beyond A the initial creep was no longer considered as linear. The initial creep rate is determined by the slope of the least-squares regression line, B.
Figure 10.--True strain versus time curves for creep of polyvinyl acetate at four stress levels; room conditions were 80° F. and 30 percent relative humidity.
Figure II.--Creep rate versus stress level curves for unmodified and thermosetting PVA at 30 percent relative humidity.
Figure 12.--Creep rate versus stress level curves for unmodified and thermosetting PVA at 65 percent relative humidity.
Figure 13.—Creep curves for thermosetting PVA at different ages from the time of casting; stress level was 2,500 p.s.i. and room conditions were 80° F. and 30 percent relative humidity.
Figure 14.--Strain reached after 2 hours versus cure time at 150° F. for thermosetting PVA; stress level was 500 p.s.i., and room conditions were 80° F. and 65 percent relative humidity.
Figure 15.--Creep and recovery curve for unmodified polyvinyl acetate. Zones of elastic and viscoelastic behavior are labeled. The stress level was 1,000 p.s.i. for the first 80 minutes; room conditions were 80° F. and 30 percent relative humidity.
Figure 16 -- Creep curves for epoxy resin extended with polysulfide; room conditions were 80° F. and 65 percent relative humidity.
Figure 17.--Creep curves for epoxy extended with polyamide; room conditions were 80° F. and 65 percent relative humidity.
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