Humidity Effects on the Creep Behavior of an Epoxy-Graphite Composite

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An exploratory investigation of humidity effects on the mechanical behavior of a unidirectional epoxy-graphite composite laminate is carried out by means of tensile creep tests. The test specimens are made from 8-ply unidirectional prepreg tape, consisting of Modmor-II graphite fibers and epoxy resin 1004. These specimens are first dried and then exposed stress-free, in a prescribed humidity-temperature environment for a 24-hr preconditioning. In order to examine the effects of humidity on matrix-dominated composite property, the creep load is applied normal to the fibers. A significant degradation in the matrix-dominant creep property is found as a result of a short-term humidity exposure. The effect is similar to that caused by temperature when humidity is held constant. Early creep rupture of the specimen under combined high temperature and humidity also was observed. It is shown that humidity effect on the matrix-dominant creep behavior may be described quantitatively by a time-humidity-temperature shift procedure when a certain creep-time limit is imposed.

Introduction

RECENT years have seen increased application of lightweight fiber-reinforced composite materials in aerospace structures. One of the most commonly used has been polymer-based, nonmetallic composites. It is well-known that polymers generally exhibit a time-dependent material behavior, and this behavior may in turn be influenced greatly by environmental factors such as temperature and humidity.

The rheological nature of polymeric materials has been studied extensively in the past. There exists a wealth of literature of both theoretical and experimental values (e.g., see Ref. 1). For most polymers, the temperature effects on their time-dependent properties may be represented analytically by the well-known method of time-temperature superposition,[1] or time-temperature shift. In much the same manner, the effects of humidity also may be described by a similar time-humidity superposition.[2] In both cases, material linearity is assumed.

Information appears concerning the rheological character of polymers that are reinforced with large amounts of rigid fibers; however, it is mostly fragmentary. In a recent account, Wang et al.[3] attempted analytically to model the time-temperature behavior of unidirectional and angle-ply epoxy-graphite composite laminated systems on the basis of Hashin's linear viscoelastic micromechanics theory.[4] Within linear response of the material, the analytical model showed reasonable agreement with the experimental results.

On the other hand, the humidity effects on the rheological behavior of fiber-reinforced polymers have been studied seriously only very recently. A number of experimental papers are now appearing in the open literature.[5-8] The purpose of the present work is to extend the investigation reported in Ref. 3. Here, we continue to study the creep behavior of the same epoxy-graphite system by means of time-humidity-temperature creep tests. However, in order to examine the matrix-dominated behavior first, we chose to test the unidirectional laminates with the creep load applied normal to the fibers. Clearly, the present effort is primarily exploration; however, it is also hoped that we can establish whether or not the aforementioned time-humidity-temperature superposition procedure is applicable in describing the combined effects of humidity and temperature. If the latter is confirmed, the analytical model developed in Ref. 3 then can be modified readily to take into account the humidity effects within the framework of linear viscoelasticity.

In the following sections, detailed test procedures and test results will be presented. It will be shown that the humidity effects on the material creep can be described by a shift in the time scale when a certain limit on the creep time is imposed.

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Experiment

General Property of Material

The material used here is the same as that studied earlier in Ref. 3, namely, 8-ply unidirectional laminates made from Modmor-II graphite fiber/resin 1004, the preparation of which has been reported in detail in Ref. 3. Here, we shall briefly record some of the relevant material properties for purpose of reference. The epoxy resin and the fibers, when tested individually under room temperature (21°C), 65% relative (room) humidity (dried specimen having had at least 24 hours exposure to room humidity), and a strain-rate of 1/6 $\min^{-1}$ were found to have the properties shown in Table 1.

The epoxy also was found to exhibit considerable nonlinear viscoelasticity when the applied load exceeds 30% of the ultimate strength, which itself is decreasing with the increase

Table 1 Properties of epoxy resin and fibers

	Resin	Fiber[a]
Modulus (10^{10}dyne/cm^2)	3.67	278.0
Poisson ratio	0.42	0.10
Strength (10^8dyne/cm^2)	5.44	204.0

[a]The properties of the fibers were found by testing 0-deg unidirectional laminates in conjunction with a rule of mixture. Since graphites are generally transversely isotropic, properties found by this method are valid only within the context of axial loading conditions (see discussions in Ref. 3).

of temperature. On the other hand, the graphite fibers essentially displayed a linear elastic behavior, within the test temperature range of 20° to 100°C. As for the composite, the unidirectional laminate was found to have a fiber volume fraction of 58%; and its longitudinal tensile strength was about 120×10^8 dyne/cm^2, whereas its transverse tensile strength was only 5.5×10^8 dyne/cm^2.

Specimen Configuration and Test Procedure

The test specimens studied in this work are cut from a unidirectional laminated plate of eight layers of prepreg tape. The thickness of the laminate averages at 0.12 cm; the overall size of the specimen is 2.5 cm wide, 18 cm long. Ends of the specimen are reinforced with fiber-glass end-tabs, leaving the effective length of the specimen at about 13 cm.

In the creep test, a standard dead-weight creep tester is used. The applied creep load is normal to the fibers, i.e., only creep strains transverse to the fibers are being measured. The entire apparatus is housed in a humidity-temperature controlled chamber, whose size is $5 \times 2 \times 2.5$ m^3. The chamber can maintain a constant temperature and humidity in the range of −55° to 95°C and 20 to 100% R.H., respectively. Before a specimen is loaded, a 0.5-cm size strain gage is mounted on the center, measuring the axial strain in the direction of the load. The specimen is then dry-cured in an oven at 120°C for at least 24 hr. The specimen so prepared is regarded as in its reference state. Twenty-four hours prior to loading, the specimen is placed stress-free in the testing chamber for continuous exposure under a predetermined humidity and temperature condition.‡ Creep load in the form of dead weights then is released onto the specimen through a remotely controlled hydraulic jack, while simultaneously the creep strain history is recorded by a graphic strain-recorder. The entire loading process is monitored from outside the chamber through a control panel and a window.

A total of 12 individual tests are performed; all are under the same dead-weight. However, owing to the slight differences in the cross-sectional area of the specimens, the effective stress varies in the range of $1.6 \pm 0.016 \times 10^8$ dyne/cm^2. Table 2 shows the conditions at which each individual test is carried out.

Perhaps it is important to note that the applied stress is about 30% of the specimen's ultimate strength at room temperature and is about 40% of that at 65°C.

Test Results

Test results are presented graphically in this report. Figures 1 and 2 show the creep strain history under various humidity conditions for temperature at 21° and 65°C, respectively. It is seen that at each given temperature the creep curves, as they are affected by various humidity, display a similar character to that displayed by curves obtained under a given humidity, but with various different temperatures.[3] Generally speaking, an increase in humidity results in an increase of the creep strain; the degree of increase is larger when temperature is higher. Clearly, the effects of humidity are compounded by the effects of temperature. There also is clear indication of nonlinear creep, especially when both the humidity and the temperature levels are high. The specimen tested under 65°C/98% R.H. failed by creep-rupture 2 hr after loading. This happened again in a repeated test at almost the same creep time.

The material relaxation moduli, calculated directly from the curves in Figs. 1 and 2, using the so-called quasielastic conversion method,[9] are shown in Figs. 3 and 4, respectively. It is seen that the relaxation moduli vary linearly with the logorithmic time only for a relatively short period; in the case for 21°C (Fig. 3), the linear portion of the curves extends to about 2 hr, whereas that for 65°C (Fig. 4) extends to only 1 hr or so. Particularly for the one curve under 65°C/98% R.H., nonlinear creep starts almost instantly, as soon as the load is applied.

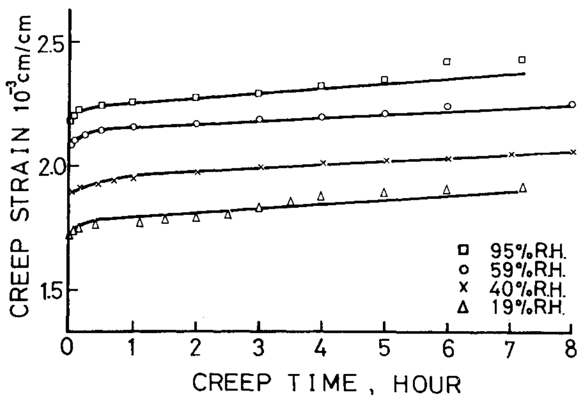

Fig. 1 Creep strain history – 21°C.

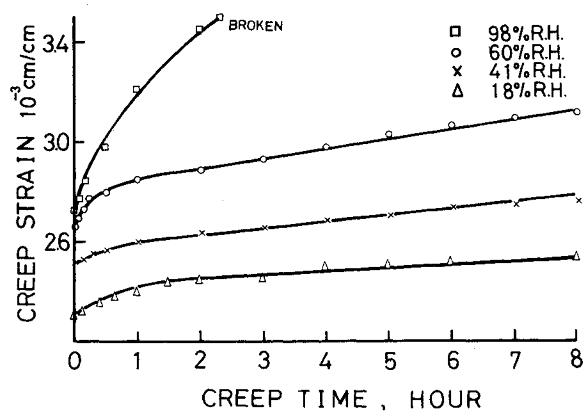

Fig. 2 Creep strain history – 65°C.

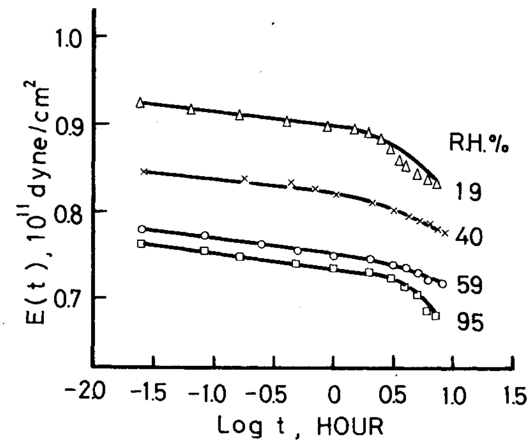

Fig. 3 Relaxation moduli – 21°C.

Table 2 Test conditions

Temp.,°C	Humidity		%	R.H.
21	19[a]	40[a]	59	95[a]
65	18	41	60	98

[a] Tests repeated once.

‡According to the latest experimental data reported,[5] for a laminate similar to that used here, full desorption of moisture content takes about 25 hr under 250°F(~110°C). On the other hand, for full absorption of moisture, under 150°F, 98% R.H., it takes more than 2 weeks, whereas under 150°F, 45% R.H., it takes about 48 days. For a 24-hr exposure, the material is only about half saturated. As for the moisture content in the specimen, a fairly accurate estimate may be obtained using the method outlined by McKague et al.[8].

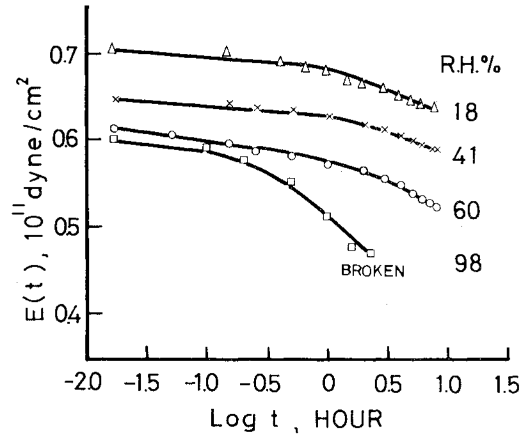

Fig. 4 Relaxation moduli – 65°C.

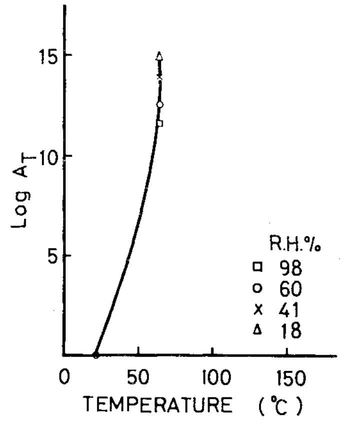

Fig. 6 Time-temperature shift factor A_T.

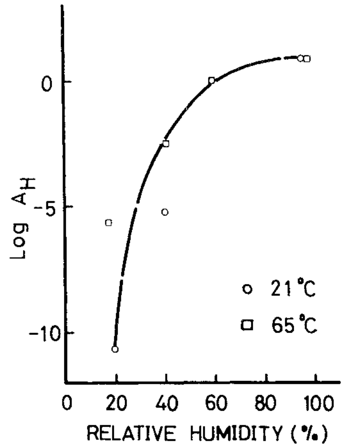

Fig. 5 Time-humidity shift factor A_H.

The exact causes for the nonlinear behavior cannot be ascertained, however. It may be a combination of the following: 1) the fact that the applied stress is already too high§ to effect linear deformation, since the applied creep stress of 1.6×10^8 dyne/cm^2 is nearly 30% the ultimate tensile strength of the specimen when tested statically under 21°C, or 40% when under 65°C; 2) that the humidity, as a swelling agent, has not reached the fullest penetration within 24-hr exposure time, and therefore it may penetrate faster when the specimen is loaded with stress (as the load may open up cracks at matrix fiber interface). Thus, as the specimen undergoes creep, the humidity penetrates deeper and weakens the rigidity of the material, resulting in nonlinear deformation. The time period in which the relaxation moduli vary linearly against the logarithmic time shortens as the temperature and/or humidity elevate (see Figs. 3 and 4).

The existence of the nonlinear creep, as it was discussed in the preceding prevents the application of the previously mentioned method of time-humidity-temperature superposition. However, in order to delineate the tangible effects of humidity and temperature on a more quantitiative basis, we shall disregard, for the moment, the nonlinear portion of the relaxation moduli shown in Figs. 3 and 4; or we shall consider only the first hour creep for all cases.¶ Within this limit, the shifting technique discussed previously then can be applied. When the temperature is held fixed, the time-humidity shift factor A_H, using 60% R.H. as reference humidity, is shown in Fig. 5. It is seen that points obtained under 21°C tend to fall side-by-side with those obtained under 65°C, although the agreement is not so close when the humidity is low. On the other hand, when the humidity is held fixed, the time-temperature shift factor A_T, using 21°C as reference temperature, is shown in Fig. 6. Again, points obtained for various humidities tend to fall together under a given temperature, although a certain amount of disagreement still exists. This points out two possible features, however; the first is that the effects of temperature and the effects of humidity are fundamentally coupled together, and thus are not independent of each other. The second is more technical, because it is possible that the specimens tested were at their various degree of moisture saturation, and that they were still

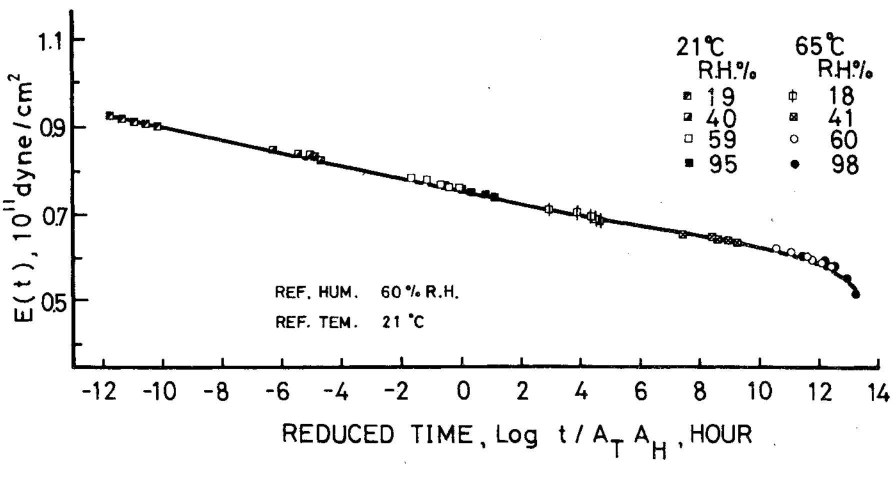

Fig. 7 Composite (master) relaxation modulus under temperature-humidity effects.

§In practical design of laminates, transverse tensile stress in individual lamina, either because of mechanical load or thermal load, may well reach beyond this level.

¶It is envisioned that, in the case of a flying aircraft (built with composites) through a rain storm, the duration of high payload/temperature/humidity would be normally 1 hr or so.

in the transient stage of moisture absorption during the creep tests. In any event, if all of these considerations are temporarily disregarded, an averaged value for A_H or A_T may be obtained (see the solid line in Fig. 5 or 6). By use of the averaged A_H and A_T, a composite master curve for the relaxation modulus of the material then can be constructed, which is shown in Fig. 7. This curve is seen to be surprisingly smooth, indicating the validity of the shift technique when only 1-hr creep is considered.

Concluding Remarks

An exploratory experimental study of the humidity effects on the mechanical behavior of an epoxy-graphite composite laminate is reported herein. Several relevant remarks are made here as a conclusion for this paper.

1) The matrix-dominant material degradation is found to be quite serious, even for a brief period of humidity exposure. Such effects cannot be ignored whenever design criteria are governed by matrix-dominant properties.

2) Like temperature exposure, humidity exposure also tends to shift the time-scale of the rheological characteristics of the material, suggesting possible utility of a linear viscoelastic model incorporating the time-humidity-temperature superposition method.

3) The present test data are rather limited; the results are relevant only to short-period humidity exposure. Effects of long-term water absorption and even dry-wet cycling are being studied in a continued effort.

4) The need for describing the nonlinear responses and/or transient environmental responses of the composite analytically also should be fulfilled.

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