# Creep Ruptures in Heterogeneous Materials

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We present creep experiments on fiber composite materials with different controlled heterogeneity. All samples exhibit a power-law relaxation of the strain rate in the primary creep regime (Andrade's law) followed by a power-law acceleration up to rupture. We discover that the rupture time is proportional to the duration of the primary creep regime, showing the interplay between the two regimes and offering a method of rupture prediction. These experimental results are rationalized by a mean-field model of representative elements with nonlinear viscoelastic rheology and with a large heterogeneity of strengths.

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The damage and fracture of materials, particularly composite materials used in naval, aeronautics, and space industry, are of enormous interest due to their economic and human cost. Despite considerable experimental [1–3] and theoretical work [4–6] on fracture, many questions have not been answered yet. Recently, statistical physicists have shown the existence of a power-law acceleration of acoustic emissions announcing the global rupture of heterogeneous materials [2,3], similar to the critical behavior of the out-of-equilibrium phase transition [6], offering a way to predict material rupture [2].

This Letter presents creep experiments on composite materials with controlled heterogeneity conducted up to rupture, which we explain using a simple model of representative elements, in the framework of fiber bundles models. Creep, also known as ''static fatigue,'' is the progressive deformation of a material under constant load. Three creep regimes are usually observed. During the primary regime, the strain rate decays as a power law with time following the application of the stress (Andrade's law) [7]. The secondary regime describes a quasiconstant deformation rate, which evolves towards the tertiary creep regime, if the stress and the temperature are high enough, during which the strain rate accelerates up to rupture.

The experiments are carried out on cross ply glass/polyester composite materials and on sheet molding compound (SMC) composites. Two types of cross angle ply laminates are fabricated, denoted $[\pm 62°]$ and $[90°/35°]$, where the angles measure the directions of the glass fibers with respect to the loading direction, with a fiber volume fraction of 75%. The SMC composites consist of a combination of polyester resin, calcium carbonate filler, thermoplastic additive, and random oriented short glass fibers, in the form of a sheet. The relatively low fiber volume fraction, about 30%, and the random filler and reinforcement distribution during processing lead to a more heterogeneous structure for the SMC compared to the cross ply composites. The $[\pm 62°]$ and $[90°/35°]$ specimens have dimensions $14 \times 100 \times 2$ mm$^3$. The SMC samples are in the form of a 120 mm barbell with 3 mm thickness. All specimens are subjected to a constant stress $\sigma$ and temperature $T$ (below the glass transition of the matrix), which were fixed to $\sigma = 15$ MPa and $T = 60\,°\mathrm{C}$ for the $[\pm 62°]$ specimens, $\sigma = 22$ MPa and $T = 60\,°\mathrm{C}$ for the $[90°/35°]$ specimens, and $\sigma = 48$ MPa and $T = 100\,°\mathrm{C}$ for the SMC. The creep tensile tests were performed using a servohydraulic mechanical testing system. Constant tensile load was applied after a transient progressive loading, and the resulting strain and acoustic emissions were recorded until rupture. Acoustic emission (AE) is a standard technique to monitor the evolution of damage in composites, due to matrix cracks, fiber matrix debounding, fiber breaks, and delaminations [8].

All 15 samples show a transient decrease of the strain rate following the application of the stress (primary creep), followed by a quasistationary regime (secondary creep), ending with an acceleration of the strain rate up to rupture (tertiary creep) (Fig. 1). The decrease of the deformation rate $d\epsilon/dt$ in the primary creep regime can be described by Andrade's law [7] $d\epsilon/dt \sim t^{-p}$, with an exponent $p \approx 1$ for all samples compatible with a universal $1/t$ decay as shown in Fig. 1(b). The deviation from Andrade's law for small time results from the transient of the first 10 sec during which the stress progressively increases up to its final and constant value.

The acceleration of the strain rate before rupture can be described by a power-law singularity $d\epsilon/dt \sim (t_c - t)^{-p'}$, with an exponent $p' \approx 1$ for all 15 samples, compatible with a universal $1/(t_c - t)$ acceleration as shown in Fig. 1(c). In Ref. [9], we also fit the strain rate and AE rate of each sample by Andrade's law in the primary regime, and by a power-law acceleration before rupture,

 

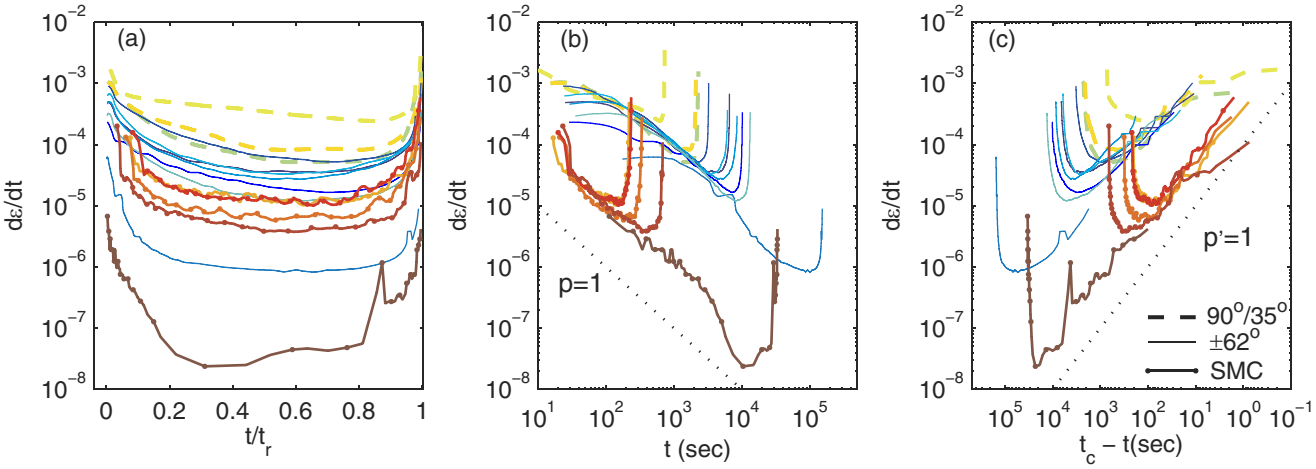


FIG. 1 (color online). Creep strain rate for all 15 samples: (a) linear time scale, with times renormalized by the rupture time $t_r$; (b) logarithmic time scale to test for the existence of Andrade's law in the primary creep; (c) logarithmic time scale in $t_c - t$ to test the time-to-failure power-law in the tertiary creep. The thick straight line in (b) [respectively, (c)] corresponds to the law $1/t$ [respectively, $1/(t_c - t)$].

letting $p$ and $p'$ free to adjust. We find that $p$ fluctuates in the range 0.3–1.3 and $p'$ varies between 0.5 and 1.1 for the 15 samples tested, with most values of $p$ and $p'$ close to 1. The critical times $t_c$ determined from the fits of the data are close to the observed rupture times for each sample. The values of $p$ and $p'$ are on average slightly larger for the SMC than for the cross ply composites, possibly due to the larger heterogeneity of the SMC or to the different values of the applied stress. However, because of the limited scaling regime used in the fits, it is difficult to decide whether this dispersion of $p$ and $p'$ results from genuine sample-to-sample fluctuations or from statistical noises and finite-size effects.

The rate of AE events, and the rate of AE energy release, exhibits a behavior similar to the strain rate (Andrade's law and power-law acceleration before rupture), with larger fluctuations and slightly different exponents, and with a few exceptions: the $[90°/35°]$ samples did not show a relaxation of AE activity during primary creep, and one $[\pm 62°]$ sample did not show any acceleration before rupture (see [9] for more details on the AE data). The difference between strain rate and AE data is not surprising as AE and strain do not reveal the same physical processes. AE measures the different types of damage developing and cascading within the sample, while strain is an integrated measure of both damage and stress redistribution within the sample. AE is more sensitive to the highest moments of the stress distribution while strain is related to the low order moments of stress. The better time resolution of the AE data compared with the strain data allowed us to observe the critical power-law acceleration of damage over a longer time range, up to 4 orders of magnitude in time [9], thus confirming the previous announcement of power laws in the tertiary creep regime which were established over more limited time scales [2,3].

There is a huge variability of the rupture time from one sample to another one, for the same applied stress, as shown in Figs. 1 and 2. These fluctuations of rupture time are expected for quenched random systems with strongly nonlinear dynamics, which are very sensitive to the heterogeneity distribution and to small variations of the applied stress. Figure 2 shows that the transition time $t_m$ between the primary creep regime and the tertiary regime, defined as the minimum of the strain rate, is proportional to the rupture time: $t_m \approx 2/3 t_r$. We also found a negative correlation between the (fitted) Andrade exponent $p$ and

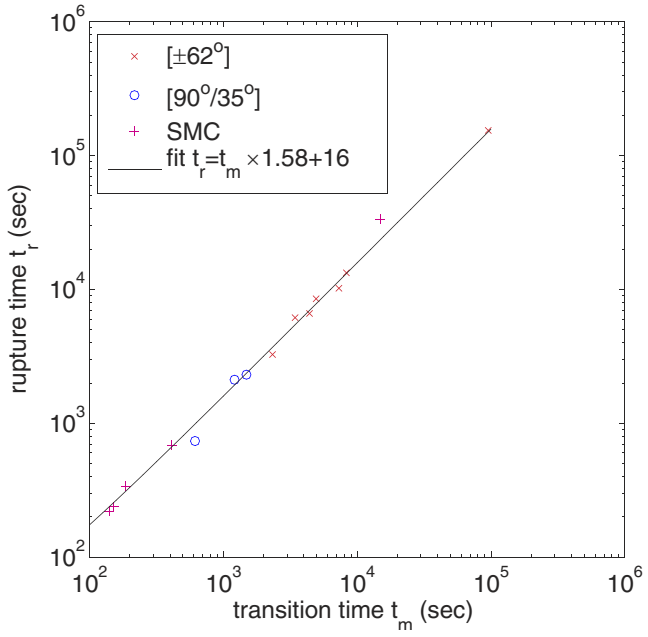


FIG. 2 (color online). Relation between the time $t_m$ of the minima of the strain rate and the rupture time $t_r$, for all samples.

the rupture time $t_r$ [9]. These observations show that damage in the primary and secondary creep regimes impacts its subsequent evolution. This suggests a way to predict the rupture time from the observation of the strain rate during the primary and secondary creep regimes, before the acceleration of the damage leading to rupture.

Creep observations have been modeled in terms of viscoelastic fibers, with deterministic dynamics and quenched disorder [10]. This model reproduces a power-law singularity of the strain rate before rupture with $p' = 1/2$ in the case of a uniform distribution of strengths [10] but does not explain Andrade's law for the primary creep. Here, we start from the model of [10] and enrich it with a more realistic rheology and heterogeneity, in order to account simultaneously for Andrade's law in the primary creep and for the power-law singularity of the strain rate before rupture. We view a composite system as made of a large set of representative elements (RE), each element comprising many fibers with their interstitial matrix. The applied load is shared democratically between all RE. This assumption has been shown to be a good approximation of the elastic load sharing for sufficiently heterogeneous materials [11]. Each RE is modeled as a nonlinear Eyring dashpot [12] in parallel with a linear spring of stiffness $E$. The Eyring rheology, which is the standard for fiber composites, consists at the microscopic level in adapting to the matrix rheology the theory of reaction rates describing processes activated by crossing potential barriers. A given RE fails when its elongation/deformation $\epsilon$ reaches a threshold. The rupture thresholds are distributed according to the cumulative distribution $P(\epsilon)$ given by $P(\epsilon) = 1 - [\epsilon_{01}/(\epsilon + \epsilon_{02})]^{\mu}$, where $\epsilon_{01}$ and $\epsilon_{02}$ are two constants with $\epsilon_{01} \leq \epsilon_{02}$. That $P(\epsilon \to 0)$ is nonzero reflects the fact that a fraction $1 - (\epsilon_{01}/\epsilon_{02})^{\mu}$ breaks as soon as the stress is applied, thus modeling a finite fraction of weak elements. The power-law distribution $P(\epsilon)$ for large $\epsilon$ is motivated by the large distribution of rupture times for the same applied stress (Fig. 2). The exponent $\mu > 1$ controls the amplitude of the frozen heterogeneity of the RE strengths.

The equation controlling the deformation $\epsilon(t)$ of each surviving RE according to the Eyring rheology is

$$\frac{d\epsilon}{dt} = K \sinh\left(\frac{\beta\sigma}{1 - P(\epsilon)} - \beta E\epsilon\right) \tag{1}$$

with the initial condition $\epsilon(t = 0) = 0$. The fraction of unbroken RE is $1 - P(\epsilon)$ and $\sigma/[1 - P(\epsilon)]$ is the stress applied on each unbroken RE. $\beta$ is proportional to the inverse temperature, $E$ is the Young elastic modulus of the RE, and $K$ is a rate coefficient.

The system defined by (1) is stable (no global rupture) if the differential equation (1) has a stationary solution $d\epsilon/dt = 0$ with $\epsilon > 0$, i.e., if the equation $[(\epsilon + \epsilon_{02})/\epsilon_{01}]^{\mu} = (E/\sigma)\epsilon$ has a nontrivial solution. This defines a threshold $\sigma^*$ below which the strain converges asymptotically to a constant and above which $d\epsilon/dt$ grows up to rupture.

In the primary regime $\epsilon \ll \epsilon_{02}$; thus $(\epsilon + \epsilon_{02})^{\mu} \approx \epsilon_{02}{}^{\mu}(1 + \mu\epsilon/\epsilon_{02})$. If the stress on the dashpot is small, we can replace sinh by exp/2. With these approximations, the differential equation (1) has the solution

$$\frac{d\epsilon}{dt} = \frac{K}{2e^{-\beta\sigma(\epsilon_{02}/\epsilon_{01})^{\mu}} + tK\beta\left(E - \frac{\mu\sigma}{\epsilon_{02}}\left(\frac{\epsilon_{02}}{\epsilon_{01}}\right)^{\mu}\right)}. \tag{2}$$

Expression (2) predicts that, if the stress is not too large, $d\epsilon/dt$ is of Andrade's form $\sim t^{-p}$, with an exponent $p = 1$ at early times, in good agreement with the average decay law shown in Fig. 1(b). For larger $\sigma$, the strain rate starts to accelerate as soon as the load is applied. Note that the observation of Andrade's power-law creep in this model does not involve any rupture of RE and is thus independent of the choice of the distribution of rupture thresholds $P(\epsilon)$.

In the tertiary creep regime, we can neglect $\epsilon_{02}$ compared with $\epsilon$. Close to rupture, for large $\epsilon$, the linear term $E\epsilon$ is small compared with $\sigma/[1 - P(\epsilon)] = \frac{\sigma}{\epsilon_{01}{}^{\mu}}(\epsilon + \epsilon_{02})^{\mu}$ if $\mu > 1$. This leads to the equation

$$\frac{d\epsilon}{dt} \approx \frac{K}{2} \exp\left(\frac{\beta\sigma\epsilon^{\mu}}{\epsilon_{01}{}^{\mu}}\right). \tag{3}$$

Its solution is, to leading logarithmic order,

$$\frac{d\epsilon}{dt} = \frac{A}{\mu}[-\ln(t_c - t)]^{(1/\mu)-1}\frac{1}{t_c - t}, \tag{4}$$

where $A = \epsilon_{01}(\beta\sigma)^{-1/\mu}$. Contrary to the primary regime, the heterogeneity of the rupture threshold is an essential ingredient for the power-law singularity before rupture, but the leading power-law term with $p' = 1$ in (4) does not depend on the exponent $\mu$ characterizing heterogeneity, in good agreement with the average acceleration law shown in Fig. 1(c). The acceleration toward rupture is due to the positive feedback of broken RE, which increases the stress and strain on the unbroken RE leading to the global rupture of the system.

Figure 3 shows the numerical solution of Eq. (1) together with the approximate analytical solutions (2) in the primary creep and (4) close to rupture, for different values of the applied stress $\sigma$. In the primary creep regime close to the rupture threshold $\sigma \approx \sigma^*$, we observe numerically an apparent exponent $p < 1$, smaller than predicted by (2), which can explain the values of $p$ fitted to the experimental data [9]. For a stress $\sigma \gg \sigma^*$, the strain rate accelerates immediately when the load is applied. For $\sigma > \sigma^*$, the apparent $p$ value decreases in the model between 1 and 0 as the applied stress increases. The duration of the primary creep also decreases with $\sigma$. The model thus explains the correlation found experimentally between the $p$ value and the rupture time [9].

In the tertiary regime, for $\sigma \gg \sigma^*$, we find numerically that expression (3) is a good approximation very close to

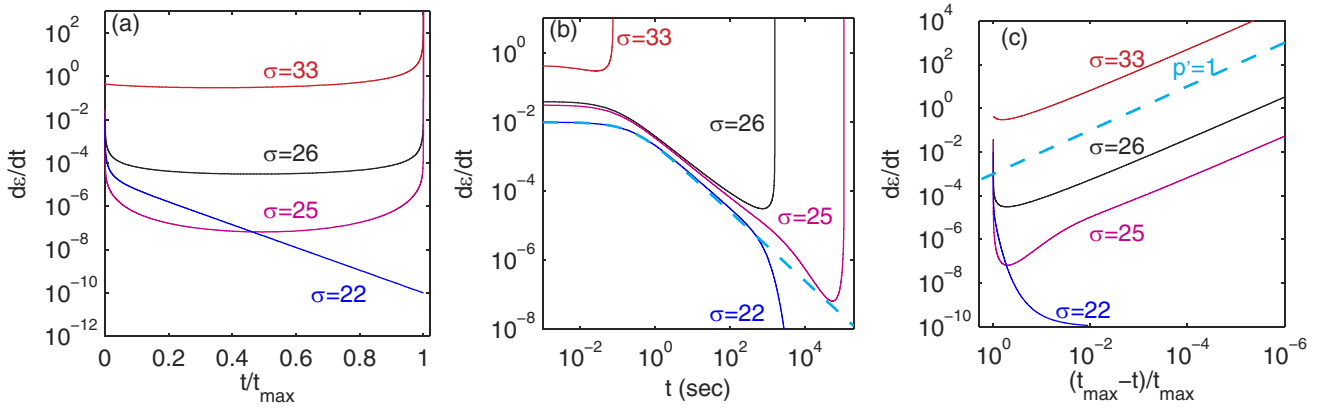


FIG. 3 (color online). Strain rate $d\epsilon/dt$ given by (1) for different values of the stress $\sigma$ (in MPa), and with parameters $E = 20$ GPa, $\mu = 1.2$, $\epsilon_{01} = 0.003$, $\epsilon_{02} = 0.015$, $\beta = 50$ GPa$^{-1}$, and $K = 10^{-5}$ sec$^{-1}$. (b) illustrates Andrade's law in the primary regime, with exponent $p \approx 1$ for $\sigma = 22$ MPa and $p \approx 0.8$ for $\sigma = 25$ MPa. The dashed line is the approximate solution (2) of (1) with $\sigma = 22$ MPa. (c) shows the power-law acceleration of $d\epsilon/dt$ before failure for $\sigma \geq 25$ MPa, with $p' \approx 1$ asymptotically. In (a) and (c) the time is normalized by the rupture time for $\sigma \geq 25$ MPa, and by the time when $d\epsilon/dt$ decreases below $10^{-14}$ for $\sigma = 22$ MPa.

rupture $t \approx t_c$. But for $\sigma \approx \sigma^*$, there is a crossover further from rupture with an apparent exponent $p' = 0.9$. This simple model thus reproduces both power laws in the primary and tertiary creep regimes, with an apparent exponent $p \leq 1$ for the primary creep, and with $p' = 1$ for the tertiary regime, except for a crossover with an apparent exponent $p'$ slightly smaller than 1.

The rupture time has a power-law singularity $\sim(\sigma - \sigma^*)^{-1/2}$ for $\sigma \approx \sigma^*$, as found previously [10] for the model with a linear dashpot, and decays exponentially for $\sigma \gg \sigma^*$ [9]. The transition time $t_m$ (minima of the strain rate) is equal to $t_c/2$. This result recovers the proportionality of $t_m$ and $t_c$ found experimentally but predicts a duration for the primary creep shorter than the observations $t_m \approx 2t_c/3$ (Fig. 2).

In conclusion, we have shown that the interactions between the RE elements together with a large heterogeneity and a simple nonlinear rheology are sufficient to explain our experiments, which exhibit Andrade's law in the primary creep regime and a finite-time singular power-law acceleration before rupture. This model replaces the need for complex memory effects (such as the integrodifferential Schapery long-memory formalism [13]) often invoked in the composite literature. A natural improvement of the model would be to relax the democratic load sharing rule as in [10] in order to introduce realistic elastic interactions. This improvement may provide a more realistic value of the constant of proportionality between $t_m$ and $t_c$ (Fig. 2).

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