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IDEAL ELASTIC, ANELASTIC, AND
VISCOELASTIC FLOW IN A METALLIC GLASS

By

A.I. Taub and F. Spaepen

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Technical Report No. 15

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| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The elastic, viscoelastic, and anelastic components of the homogeneous strain response of the metallic glass Pd ₈₂ Si ₁₈ to an applied stress have been examined. The elastic response is fully reversible, instantaneous, and linear. The measured elastic modulus and temperature dependence are $E = 84 \pm 8$ GPa and $d(\ln E)/dt = (-3.2 \pm 0.6) \times 10^{-4} \text{ C}^{-1}$. This viscoelastic flow is nonrecoverable, and, if the configuration remains constant, is characterized by a constant strain rate. This strain rate varies linearly with the stress in the low stress regime ($\gamma < 300$ MPa). | | |

becoming nonlinear for higher stresses. For isoconfigurational flow, the strain rate has an Arrhenius-type temperature dependence with an activation energy of -200 ± 15 kJ/mole, independent of stress and thermal history. The magnitude of the strain rate is strongly dependent on the degree of structural relaxation and therefore on thermal history. During isothermal annealing, the viscoelastic strain rate varies inversely with time.

The anelastic response is a transient that at 500 K contributes to the flow for approximately 50 hours after a stress increase and is fully recovered upon stress reduction. A spectrum of exponential decays is required to model this flow component. The anelastic strain varies linearly with the magnitude of the stress change over the entire stress range tested, $[\gamma_A/\Delta T = (8.0 \pm 0.80) \times 10^{-6} \text{ cm/cm-MPa}]$.

IDEAL ELASTIC, ANELASTIC, AND VISCOELASTIC FLOW IN A METALLIC GLASS

A.I. Taub and F. Spaepen*

INTRODUCTION

When a material is subjected to an applied stress, its strain response generally consists of several components, each differing by its dependence on time, by the degree to which it is recovered upon removal of the stress, and by the linearity of the response.⁽¹⁾ Glassy metallic alloy systems have been reported to exhibit at least four of these strain components:

1. Ideal elasticity (recoverable, instantaneous, linear stress-strain)^(2,3)
2. Anelasticity (recoverable, time-dependent, linear stress-strain)⁽⁴⁻⁸⁾
3. Viscoelasticity (permanent, time-dependent, linear stress-strain rate)^(5,6,9,10)
4. Instantaneous plasticity (permanent, instantaneous, nonlinear stress-strain)⁽¹¹⁾

In this study, an amorphous Pd-based alloy was tested in the homogeneous flow regime. This eliminates the strain contribution of the instantaneous plastic flow mechanism.^(12,13) For each of the three remaining strain components, the time dependence, recoverability, and linearity are examined in detail. The effect of structural relaxation on each type of strain is also discussed.

RESULTS

The tensile creep flow of amorphous Pd₈₂Si₁₈ wires was measured by the technique described previously.⁽⁹⁾ Both as-quenched and preannealed specimens were tested. The preanneals served to stabilize the structure of the specimens, thus permitting isoconfigurational testing. For preannealed specimens that were to undergo stress reductions during the creep test, load cycling was conducted during the anneal in order to eliminate any structural relaxation that might be associated with load removal during the test.⁽¹⁴⁾

Figure 1a shows a typical strain vs time plot obtained for a preannealed specimen after a stress increase. The preanneal in this case was 325 hours at 500 K. The instantaneous elongation γ_E is characterized by ideal elasticity. The steady-state strain γ_P , established after approximately 50 hours, is viscoelastic. The anelastic strain contribution γ_A is the transient flow that occurs before the steady-state condition is established.

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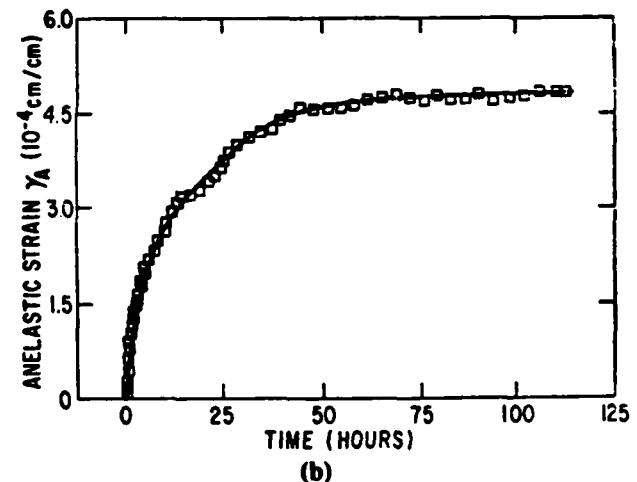
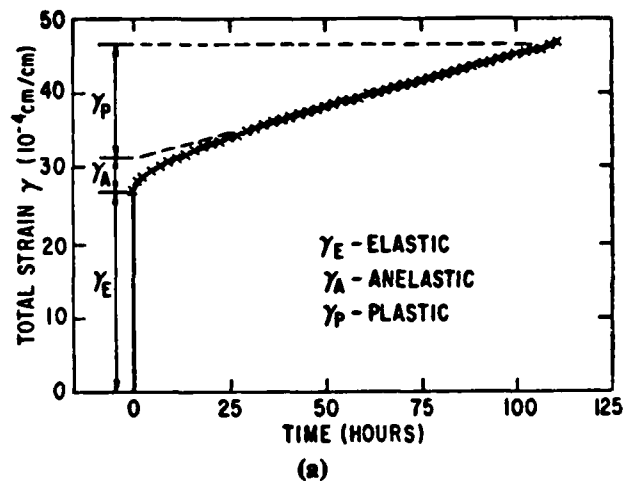


Figure 1. Strain response to equivalent shear stress increase from 105 to 156 MPa. Sample preannealed at testing temperature of 500 K for 343 hours. Symbols \times and \square are actual data. Solid lines are least-square curve fits using the five exponential decay formulation described in the text.

- (a) Total equivalent shear strain ($\gamma = \sqrt{3}$ ϵ -uniaxial tensile strain)
- (b) Equivalent anelastic shear strain γ_A , obtained by subtracting the ideal elastic and viscoelastic strains.

Ideal Elasticity

The elastic response of specimens preannealed at 500 K for 325 hours was measured at 293, 424 and 500 K. In all cases, the response was instantaneous, completely reversible, and linear.

The tensile elastic modulus (i.e., Young's modulus) at 293 K was determined to be 84 ± 8 GPa. This is in good agreement with Davis' (11) value for $\text{Pd}_{80}\text{Si}_{20}$ of 88 GPa.

Using the values of the moduli determined at all three temperatures and assuming a linear temperature dependence, we found that $d(\ln E/dT = (-3.2 \pm 0.6) \times 10^{-4} \text{C}^{-1}$. This is to be compared with the Ferry and Pritchett (15) dynamic measurement of $-2.9 \times 10^{-4} \text{C}^{-1}$.

Structural changes in the amorphous state have been shown to affect the elastic stiffness of metallic glasses. (2) Increases in modulus approaching 10%, relative to the as-cast condition, have been reported for many systems annealed near the glass transition temperature. (3) In our tests, the 325-hour preanneal at 500 K was sufficient to stabilize the structure, and no changes in modulus were observed during the loading and unloading tests.

Anelasticity

Figure 1b shows the anelastic component of the total strain of Figure 1a, obtained by subtracting the ideal elastic and viscoelastic contributions. The time-dependent, transient nature of the flow is evident. Additional anelastic flow is not resolvable after approximately two days.

Complete recoverability is a requirement for true anelastic behavior. In Figure 2, the results of a test of this condition are examined by observing the response to a stress cycle from 35 to 71 to 35 MPa. The entire response is plotted in Figure 2a. The elastic response is seen to be completely and instantaneously recovered on stress reduction ($\gamma_E = -\gamma_E'$). In Figure 2b, only the anelastic contribution is shown. Complete recoverability of this strain response is verified ($\gamma_A = -\gamma_A'$).

The linearity condition for true anelastic flow requires that the total anelastic strain, after the complete decay of the transient, be directly proportional to the magnitude of the stress change. To check this requirement, a creep sample was subjected to sequential stress increases at 500 K, and the anelastic strain was determined in the manner illustrated in Figure 1. In Table 1, the observed anelastic strain is listed for the stress increments, and a linear relation is verified. $\gamma_A/\Delta\tau = (8.0 \pm 0.80) \times 10^{-6} \text{ cm/cm-MPa}$, where τ is the equivalent shear stress $\tau = \sigma/\sqrt{3}$ and σ is the uniaxial tensile stress.

A corollary of the linearity postulate is the superposition (additivity) of the responses. (1) In another load cycle experiment, this principle was checked. The stress was raised in two steps from

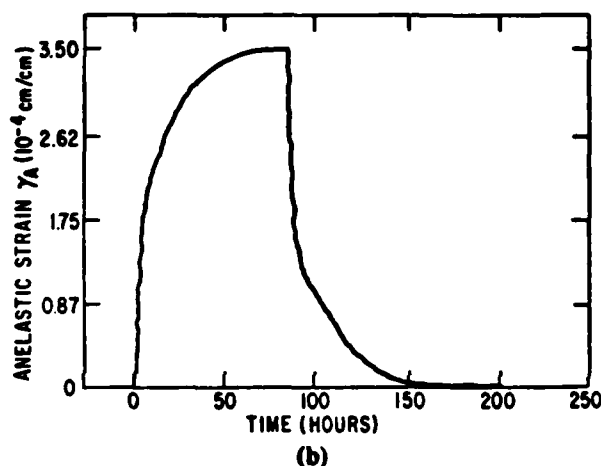
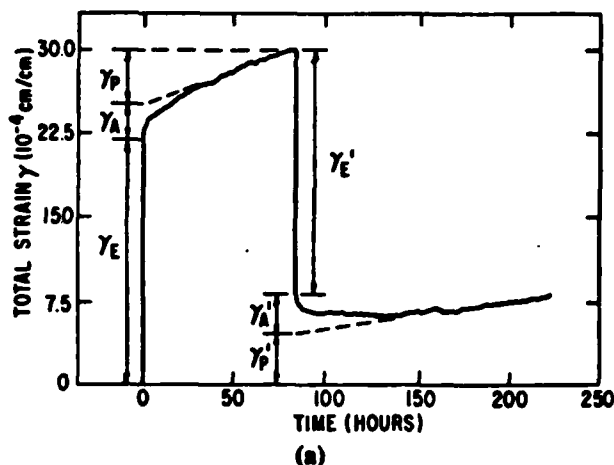


Figure 2. Strain response to stress cycle from 35 to 71 to 35 MPa. Sample preannealed at testing temperature of 500 K for 575 hours.

(a) Total strain
(b) Anelastic strain.

Table 1
TOTAL ANELASTIC STRAIN
FOR VARIOUS STRESS INCREASES*

| Equivalent Shear Stress Range $\tau = \sigma/\sqrt{3}$ (MPa) | Total Anelastic Equivalent Shear Strain $\gamma_A = \sqrt{3} \epsilon_A$ (10^{-4} cm/cm) | $\gamma_A/\delta\tau$ (10^{-6} MPa $^{-1}$) |
|---|--|--|
| 54 to 105 | 4.0 ± 0.3 | 7.8 ± 0.6 |
| 105 to 156 | 4.7 ± 0.3 | 9.0 ± 0.6 |
| 156 to 207 | 4.2 ± 0.3 | 8.1 ± 0.6 |
| 207 to 259 | 4.2 ± 0.3 | 8.1 ± 0.6 |
| 259 to 310 | 3.6 ± 0.7 | 7.2 ± 1.2 |
| 310 to 363 | 3.8 ± 0.3 | 7.2 ± 0.6 |
| 363 to 383 | 1.7 ± 0.3 | 8.4 ± 1.8 |

*Specimen preannealed at testing temperature of 500 K for 343 hours.

35 to 71 to 106 MPa. The associated anelastic strains were $(3.5 \pm 0.2) \times 10^{-4}$ cm/cm and $(3.8 \pm 0.3) \times 10^{-4}$ cm/cm. The stress was then reduced in one step to 35 MPa, with a corresponding anelastic recovery of $(7.0 \pm 0.3) \times 10^{-4}$ cm/cm. The superposition of the responses was thus verified.

Although several investigators have examined the effect of structural changes on the anelastic relaxation spectrum of metallic glasses,^(2,4) no systematic investigation of the effect of structural change on the anelastic creep response of these glasses has been reported. We observed no change in the anelastic response of the preannealed specimens during the testing, indicating that the 325-hour preanneal at the testing temperature sufficiently stabilized the structure. It is possible that the anelastic creep response changes during the early stages of annealing, but we have not made observations in this regime.

Viscoelasticity

The viscoelastic flow exhibited by well-annealed samples (i.e., no structural relaxation occurring during the test) is characterized by a constant strain rate. Referring to Figure 1, the viscoelastic component is shown to contribute throughout the test, although constant strain rate steady-state flow is not fully established until after the decay of the anelastic transient.

Viscoelastic flow must, by definition, produce a permanent strain. The test shown in Figure 2a can be used to check this requirement. The total viscous flow γ_p , up to 84 hours (when the stress was reduced), is $(5.0 \pm 0.3) \times 10^{-4}$ cm/cm. The observed permanent set γ_p' is $(5.7 \pm 0.5) \times 10^{-4}$ cm/cm, verifying nonrecoverability of the flow.

The linearity of the viscoelastic stress-strain rate relation has been discussed elsewhere.⁽¹⁶⁾ A transition from linear to nonlinear behavior was shown to be an inherent property of metallic glasses. For $\text{Pd}_{82}\text{Si}_{18}$ specimens preannealed and tested under the same condition as those discussed in this study, the limiting condition for linear flow was established at an equivalent shear stress $\tau = \sigma/\sqrt{3} = 300$ MPa, where σ is the uniaxial tensile stress.

The temperature dependence of the isoconfigurational viscoelastic strain rate has been measured. The results are reported in terms of the viscosity $\eta = \tau/\dot{\gamma}_p$ (see Figure 3). To stabilize the structure, the specimens were preannealed under an argon atmosphere with no applied stress. The annealing temperatures and times are indicated. These preanneals enabled us to test the specimens

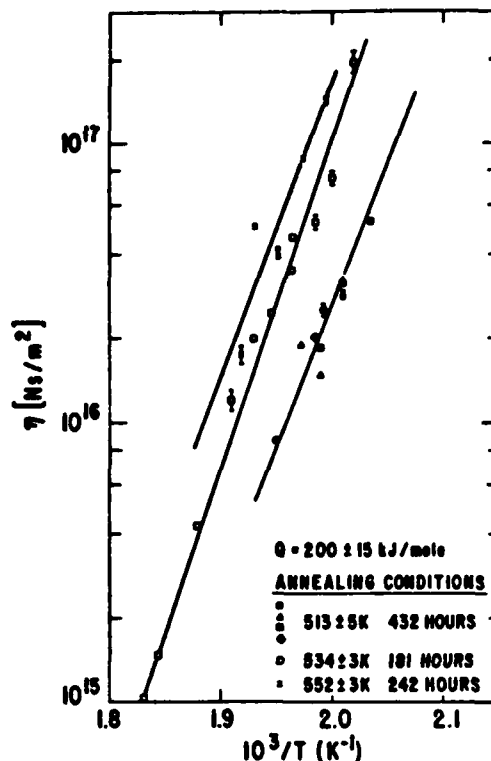


Figure 3. Isoconfigurational viscosities obtained after the indicated annealing treatments.

in the temperature range 490 to 525 K without observing additional structural relaxation. The absence of structural change was verified in each case by returning to the initial testing temperature at the end of the temperature cycle and observing no significant change from the initial value of the viscosity. The activation energies found for the isoconfigurational viscosity of these specimens is 200 ± 15 kJ/mole, in agreement with other measurements for specimens preannealed at lower temperatures.^(9,17) Tests at different stresses also showed no change in the activation energy.

Although the activation energy for isoconfigurational flow is stress and thermal history independent, the magnitude of the viscoelastic strain rate is highly dependent on the degree of structural relaxation, and therefore on thermal history. Previous investigations^(9,17) have shown that the viscosity can be changed by many orders of magnitude, even when annealing at temperatures as low as $T_g - 200$. Figure 4 shows viscosity-time histories for specimens tested under different stresses at 500 K from the cast state (curves A, B, and C). The linear increase of viscosity with time has previously been observed.^(17,18) Note that for the stress range tested ($39 < \tau < 155$ MPa), the viscosity annealing kinetics are stress-independent. Further-

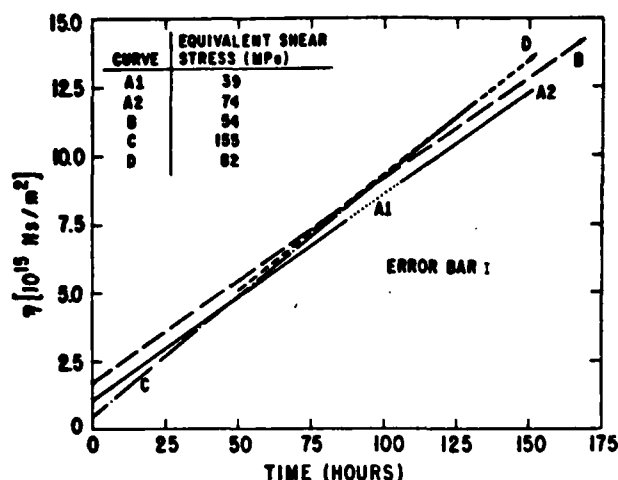


Figure 4. The viscosity as a function of annealing time for specimens tested at the indicated stresses. Samples A, B, and C were creep tested from the as-cast state. The stress on sample A was increased after 80 hours of testing. Sample D was preannealed as described in the text. The origin for sample D was shifted to $t = 50$ hours.

more, there is evidence that subsequent annealing kinetics are not affected by previous thermal history. Curve D is for a specimen that was sequentially preannealed at 424 K for 225 hours, 444 K for 175 hours, 466 K for 152 hours, and 487 K for 240 hours. The specimen was then creep tested at 502 K. During the test, the viscosity was found to increase linearly with time from an initial value of 5×10^{15} Ns/m². In Figure 4, the viscosity-time plot for this test is shown with the origin shifted to coincide with the other curves. The subsequent annealing kinetics are seen to be the same as those for the as-cast specimens, implying that the kinetics of structural relaxation are independent of the thermal history.

For certain properties, such as the Curie temperature,⁽¹⁹⁾ a "cross over effect" is observed during structural relaxation, indicating a thermal history memory. On the other hand, the viscosity does not appear to exhibit this property.

DISCUSSION

Anelastic flow in many crystalline alloys can be characterized as a single process with the general form of an exponential decay: $\gamma_A = \gamma_A^0(1 - e^{-t/\tau})$.⁽¹⁾ We attempted to fit the flow observed in these tests with this relation combined with a viscoelastic term $\gamma(t) = \gamma_A^0(1 - e^{-t/\tau}) + \dot{\gamma}_p t$. A typical least-squares fit, using γ_A^0 , τ , and $\dot{\gamma}_p$ as free parameters, is shown in Figure 5. Only the anelastic strain is shown, the viscoelastic strain $\dot{\gamma}_p t$ having been subtracted from the total strain. As a result of this

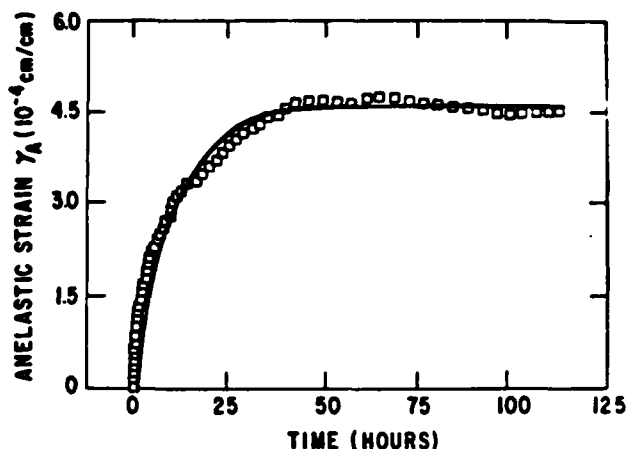


Figure 5. The equivalent anelastic shear strain computed from the data of Figure 1a, using a single exponential decay formulation. The symbols are the actual data points, from which the fitted viscoelastic strain has been subtracted. The solid line is the least-squares fit.

subtraction, the anelastic data peaks at approximately 70 hours and then drops. This is physically unrealistic, and is only an artifact of the fit value for the viscoelastic strain rate $\dot{\gamma}_p$ being higher than the actual value. In addition, the data starts higher than the fitted curve and then crosses it twice before peaking. Similar systematic deviations were observed in every case examined in this manner. This incompatibility of the data with a single process exponential decay is an indication of the existence of an anelastic site spectrum. The existence of such a spectrum in the more common glassy systems is well established.⁽²⁰⁾ Recent work on metallic glasses has shown that anelastic flow in these materials is also governed by a spectrum.^(8,15) The anelastic flow can then be expressed as $\gamma_A = \sum_j \gamma_A^j (1 - e^{-t/\tau_j})$, where the sum is replaced by an integral for the case of a continuous spectrum. Fitting experimental data by an exact method to such a relation is difficult, and one must resort to an approximation technique.⁽²¹⁾ We chose the finite spectrum approach, employing a least-squares fit to the prefactor of five exponential decays, with predetermined time constants spaced by a factor of three ($\tau_j = 0.2, 0.6, 1.8, 5.0,$ and 17 hours). The smallest time constant is limited by the data acquisition rate, one point every 0.1 hour. The decay with the largest time constant, 17 hours, reaches 0.95 of its final value after 50 hours. From the recovery tests, it is known that additional anelastic flow is undetectable after this time. Therefore, this spectrum spans the necessary time constants. Performing a least-squares fit to the data

with this relation combined with a viscoelastic term $\gamma(t) = \sum_j \gamma_j (1 - e^{-t/\tau_j}) + \dot{\gamma}_p t$, and using γ_j and $\dot{\gamma}_p$ as free parameters, we produced an excellent fit to the data (see the solid lines in Figure 1). Note that the fit value for the viscoelastic strain rate $\dot{\gamma}_p$ is reasonable since it does not produce an artificial peak in the anelastic strain. This method provides an analytic expression from which the total anelastic flow can be computed. However, fitting the data with a finite sum of exponentials whose time constants are arbitrarily predetermined, yields a nonunique solution, and therefore no physical significance could be attributed to the weights of the resulting spectrum.^(1,21)

CONCLUSIONS

The homogeneous strain response of amorphous $\text{Pd}_{82}\text{Si}_{18}$ to an applied stress consists of three components: elastic, anelastic, and viscoelastic strains.

The elastic response is fully reversible, instantaneous, and linear. The tensile elastic modulus at 293 K was determined to be 84 ± 8 GPa. The temperature dependence is $d(\ln E)/dT = (-3.2 \pm 0.6) \times 10^{-4} \text{C}^{-1}$.

The anelastic response is a transient that follows stress increases and is fully recovered upon stress reduction. For a $\text{Pd}_{82}\text{Si}_{18}$ specimen, preannealed and tested at 500 K, the anelastic transient contributes measurably of the flow for approximately 50 hours. The transient cannot be described by a single process exponential decay. However, a sum of exponential decays spanning a spectrum of time constants from 0.2 to 17 hours provides an excellent fit to the data.

The total anelastic strain, after the complete decay of the anelastic transient, varies linearly with the magnitude of the stress change over the entire stress range tested: $\gamma_A/\Delta\tau = 8.0 \pm 0.8 \times 10^{-6} \text{MPa}^{-1}$. The superposition principle was shown to apply to anelastic flow.

The isostructural viscoelastic flow is nonrecoverable and characterized by a constant strain rate. The activation energy for isoconfigurational flow was found to be 200 ± 15 kJ/mole, independent of the annealing temperature used to stabilize the structure. When structural relaxation occurs, the viscoelastic strain rate changes by many orders of magnitude. The strain rate has been observed to vary inversely with time. The rate of decrease was found to be both stress and thermal history independent.

The viscoelastic strain rate-stress relation is linear only in the low stress regime, a transition to nonlinear behavior occurring at approximately

300 MPa. The anelastic strain, on the other hand, remains linear over the entire stress range tested (0 to 383 MPa).

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