Viscoelastic behaviour in indium tin alloys over a wide range of frequency and time

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by

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ABSTRACT

Experimental studies of dynamic and transient viscoelastic response were conducted at 24°C on γ InSn alloy, and high frequency studies were conducted to extend the frequency range for eutectic InSn. The experiments were conducted in torsion using an instrument capable of determining viscoelastic properties over more than ten decades of time and frequency. The damping, tan δ, followed a ν^{-n} dependence, with $n \approx 0.2$, over many decades of frequency ν. This dependence corresponds to a stretched exponential relaxation function, and is attributed to a dislocation-point defect mechanism. It is not consistent with a self-organized criticality dislocation model which predicts tan δ $A\nu^{-2}$. Dislocation damping in metals is relevant to development of high damping metals, the behaviour of solders and of support wires in Cavendish balances.

§1. INTRODUCTION

Viscoelastic phenomena bring to mind polymers since such effects are very pronounced in many polymeric materials (Ferry, 1979). Specifically large viscoelastic effects (damping as quantified by the loss tangent, tan δ, from 0.1 to 1 or more) are common in polymers at ambient temperature. By contrast in structural metals such as steel, brass, and aluminum, viscoelastic effects are usually small: tan δ is 10^{-3} or less (Zener, 1948, Nowick and Berry, 1972); some aluminum alloys may exhibit very small loss, e.g. type 6061-T6, with tan $\delta = 3.6 \times 10^{-6}$ (Duffy, 1990). Dynamic viscoelasticity is referred to as internal friction, and recoverable viscoelasticity as anelasticity. The loss angle δ is the phase angle between stress and strain during oscillatory loading.

Viscoelasticity in metals is of interest for a variety of reasons. For example, materials with a high loss tangent are of use in damping vibration in structures and vehicles. Since structural metals have low damping, polymer layers are of use as attached layers in providing damping for bending vibration. Polymer layers do not damp compressional vibration well and they have drawbacks in aggressive environments. Stiff materials with high damping would be useful in such situations. Another example consists of low melting point alloys, including InSn, which are used in soldering (Hwang, 1991). Such alloys can exhibit substantial viscoelasticity. The effective performance of these solders in electronic devices is related to their viscoelastic behaviour (creep);

flow or cracking of a solder joint can lead to failure of the device containing that joint. Finally, systematic errors in the fundamental measurement of the gravitational constant have recently been attributed to viscoelasticity at low frequency in the suspension wires of the Cavendish torsion balance used in the measurements (Maddox, 1995; Quinn, *et al.* 1992, 1995).

Substantial viscoelastic response in metals is commonly but not exclusively associated with high homologous temperature $T_H > 0.5$ in which

$$T_H = \frac{T}{T_{melting}}, \qquad (1)$$

with T as the absolute temperature. Structural metals are in the regime $T_H > 0.5$ only at elevated temperature, but for some elements such as Cd, In, Pb, and Sn and alloys of low melting point, a high homologous temperature occurs at room temperature. Lead is popularly viewed as a high-loss metal, but it exhibits a relatively small peak $\tan\delta = 0.015$ in bending (Kamel, 1949) and $\tan\delta = 0.005$ to 0.016 in the audio range in torsion (Cook and Lakes, 1995). Cadmium exhibited a substantial loss tangent of 0.03 to 0.04 over much of the audio range of frequencies. Eutectic indium tin alloy which exhibited substantial damping exceeding 0.1 below 0.1 Hz (Brodt, Cook and Lakes, 1995), was used to make a composite exhibiting high stiffness and high damping (Brodt and Lakes, 1995).

The current investigation was designed to explore viscoelasticity in several alloys over a wide range of frequency and time, and to examine implications for dislocation processes at small strain in metals.

§2. METHODS

InSn alloy was obtained in a eutectic composition 52% Sn, 48% In; the γ-phase of InSn was prepared by melting the constituents in proportions 83.4 wt% Sn, 26.6 wt% In. Metals were obtained from Johnson Matthey Alfa. Ingots of metal were cut into segments and cast under argon into cylindrical form 3.1 mm in diameter. The γ-phase InSn, thought to be formed by a peritectic reaction (Hansen and Anderko, 1958) has a hexagonal crystal structure in contrast to the body centered tetragonal structure of Sn. Specimen ages (following casting) at the beginning of the tests were: eutectic InSn 192 days and γ InSn, 97 days.

Viscoelastic measurements were performed in torsion at 24°C using apparatus of Chen and Lakes (1989) as modified by Brodt, *et al.* (1995). This device (Fig. 1) permits measurements over an unusually wide range of time and frequency, under isothermal conditions. Such capability is particularly useful in composites and other materials which are not thermorheologically simple. The wide frequency range is obtained by eliminating resonances from the devices used for loading and for displacement measurement, by minimizing the inertia attached to the specimen, and by use of a geometry giving rise to a simple specimen resonance structure amenable to simple analysis. Higher frequencies (10^4 to 10^5 Hz) became accessible following design modifications (Quackenbush, 1995) permitting study of higher harmonic modes. Torque (sinusoidal for dynamic studies and step function for creep studies) was produced electromagnetically by a Helmholtz coil acting upon a high intensity neodymium iron boron magnet at the specimen free end. Angular displacement was measured via laser light reflected from a small mirror upon the magnet to a split-diode light detector. At resonant frequencies, $\tan\delta$ was inferred from the width of the dynamic compliance curve or from free decay of vibration, and in the subresonant domain, from the phase angle between torque and angle. Calibrations were performed using the well-characterized 6061-T6 aluminum alloy (G = 25.9 GPa, $\tan\delta \approx 3.6 \times 10^{-6}$) (Duffy, 1979). The surface shear strain at 1 Hz was 6.7×10^{-6} for eutectic InSn, and 1.1×10^{-5} for γ In Sn. Load level was intentionally varied in tests of linearity at the higher frequencies.

§3. RESULTS AND DISCUSSION

§3.1 Phenomenology and constitutive

Viscoelastic properties of eutectic and γ- InSn over wide ranges of time and frequency are plotted in Figure 2; also shown for comparison are the properties below 20 kHz of eutectic InSn presented earlier by Brodt, *et al.* (1995).

The γ-InSn exhibited less damping than the eutectic composition of InSn by as much as a factor of 7. Moreover, γ-InSn exhibited a smaller damping, by up to a factor of three, than Sn alone; also a smaller damping than In alone. Tan δ followed a ν^{-n} dependence over many decades for both alloys. The value of n was 0.28 for eutectic InSn (above 0.01 Hz), and 0.22 for γ- InSn (below 10 kHz). Similar ν^{-n} behaviour was seen at low frequency in pure In ($n \approx 0.2$) and Cd ($n \approx 0.086$) over the narrower frequency range explored by Cook and Lakes (1995), as well as in other alloys (unpublished). Neither stiffness nor tan δ at higher frequencies depended significantly on strain level, from 2×10^{-6} up to more than 2×10^{-5}, hence the behaviour may be regarded as linear.

The observed ν^{-n} dependence of damping, with $n < 1$, may be contrasted with the Debye form

$$\tan\delta = \Delta \frac{\omega\tau}{1+\omega^2\tau^2} \qquad (2)$$

with τ as a relaxation time and $\omega = 2\pi\nu$, with ν as frequency and Δ as a constant. Debye peaks cover about one decade of time or frequency, and are associated with single exponentials in the creep or relaxation behaviour. At frequencies well above the Debye peak, tan $\delta \approx \nu^{-1}$; observed behaviour does not follow this form. A distribution of relaxation times may be used to model experimental data but in the absence of evidence of mechanisms of this form, we shall not pursue this.

The stretched exponential form of the relaxation modulus G(t),

$$G(t) = (G_o - G_\infty)e^{-(t/\tau)^\beta} + G_\infty \qquad (3)$$

has been observed in many materials (Kohlrausch, 1847; Ngai, 1979; Palmer, Stein, Abrahams, and Anderson, 1984). Some dielectric systems, with broad asymmetric loss peaks, require a more general model (Jonscher, 1977). Here $0 < \beta < 1$, and G_o and G_∞ are constants. The case $\beta = 1$ corresponds to the Debye relaxation. Dynamic behaviour corresponding to this relaxation function can be obtained analytically for $\beta = 0.5$ (Williams and Watts, 1970) but not for general values of β. In viscoelastic liquids, $G_\infty = 0$, and tan $\delta \approx \nu^{-n}$; in viscoelastic solids, $G_\infty > 0$ and tan δ forms a broad peak with tan $\delta \approx \nu^{-n}$ for frequencies well above the peak, demonstrated numerically. The stretched exponential is a reasonable model for much of the behaviour observed here.

§3.2 Viscoelastic Mechanisms

Several physical mechanisms give rise to viscoelastic damping in metals (Zener, 1948, Nowick and Berry, 1972). Viscous slip at the grain boundaries (Zener, 1941, Kê, 1947) as well as rearrangement of pairs of atoms in an alloy (Zener, 1947) give rise to peaks in tan δ which approximate the Debye form, Eq. 2. These are operative at high homologous temperature. The dominance of the ν^{-n} dependence of tan δ suggests that the damping due to grain boundary or Zener pair rearrangement is minor compared to damping due to other sources, presumably dislocations, as described below.

High temperature background damping in metals occurs over a wider range of frequency or temperature. It is thought that the background is caused by a combination of thermally activated dislocation mechanisms. Dislocation mechanisms were examined in detail by Granato and Lücke (1956). The concept of a pinned dislocation loop oscillating under the influence of an applied stress leads to two components of loss, one which depends on frequency and attains a peak in the MHz range, and one which is a hysteresis type independent of frequency but dependent on strain. It is thought (Cagnoli, Gammaitoni, Marchesoni, and Segoloni, 1993) that such a model cannot account for damping at very low frequency.

The dependence of the background loss upon temperature and frequency has been examined by Schoeck, Bisogi, and Shyne (1964). They considered a thermally activated dislocation-point defect mechanism. If the dislocation experiences a restoring force represented by q, the damping follows a Debye peak in angular frequency $\omega = 2\pi\nu$, with ν as frequency:

$$\tan\delta = \frac{\gamma\Lambda G b}{q}\,\frac{\omega\tau}{1 + \omega^2\tau^2}\,. \tag{4}$$

in which G is the shear modulus. The dislocation has length Λ and Burgers vector magnitude b; γ is a geometrical orientation factor of order of magnitude 0.1. $\tau = \frac{1}{pq}\exp\frac{U_0}{kT}$ in which U_0 is an activation energy, T is the absolute temperature (in K), k is Boltzmann's constant and p depends on temperature only. If there is no restoring force q, $\tan\delta \quad \nu^{-1}$. For a *distribution* of activation energies, the following can be obtained.

$$\tan\delta \quad \nu^{-n}. \tag{5}$$

This is the form followed by both alloys over a wide range of frequency. Since it seems fortuitous that similar distributions occur in a variety of metals, it is natural to consider mechanisms which give rise to the observed behaviour without the assumption of special distributions.

The form $\tan\delta \quad \nu^{-n}$ follows from a stretched exponential relaxation, Eq. 2, which arises naturally in many complex materials with strongly interacting constituents. In particular, Palmer, *et al.*, (1984) showed that stretched exponential relaxation arises from relaxation in hierarchical stages such that the constraint imposed by a faster degree of freedom must relax before a slower degree of freedom can relax. The underlying feature of theories giving rise to such relaxation is the generation of a scale invariant distribution of relaxation times (Klafter and Schlesinger, 1986). Stretched exponentials in slow relaxation are so widespread as to be considered "universal" (Ngai, 1979), perhaps because they represent a probability limit distribution (Scher, Schlesinger, and Bendler, 1991).

Recent experimental study of CuBe alloy (Quinn, *et al.* 1992, 1995) disclosed $\tan\delta \quad \nu^{-2}$ for low frequency damping at a relatively high strain exceeding 10^{-4}; similar behaviour in aluminum alloy and in tungsten fibers suggested the mechanism responsible for damping in CuBe may not be unique to that material. Moreover, recent theoretical development (Cagnoli, *et al.*, 1993) assuming self-organized criticality of stick-slip dislocation processes gives $\tan\delta \quad \nu^{-2}$. The theory is nonlinear, with frequency dependent and strain dependent expressions in multiplicative form, therefore it cannot account for results of the present study at small strain.

Since several metals exhibit $\tan\delta \quad \nu^{-n}$ with $n < 1$ at small strain, it is tempting to consider the possibility of a 'universal' dislocation mechanism based on scale invariance. Since fewer mechanical systems than dielectric systems have been studied over wide ranges of frequency, a final decision on dislocation mechanisms awaits further study. In particular, the present results in combination with those of Quinn, *et al.* (1992, 1995) suggest different frequency dependence at small and large strain, and possibly different mechanisms.

§4. CONCLUSIONS

Tan δ followed a ν^{-n} dependence over many decades of frequency for these alloys. Results are consistent with a stretched exponential relaxation function, and are attributed to a dislocation-point defect mechanism. Results are not consistent with a self-organized criticality dislocation model which predicts $\tan\delta \quad A\nu^{-2}$.

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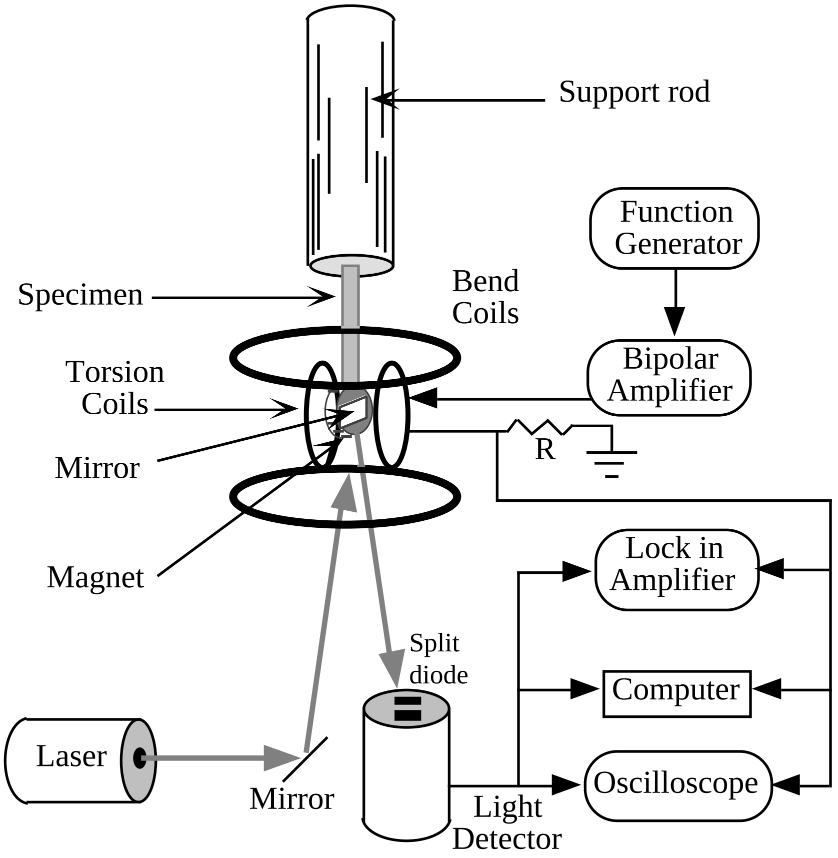
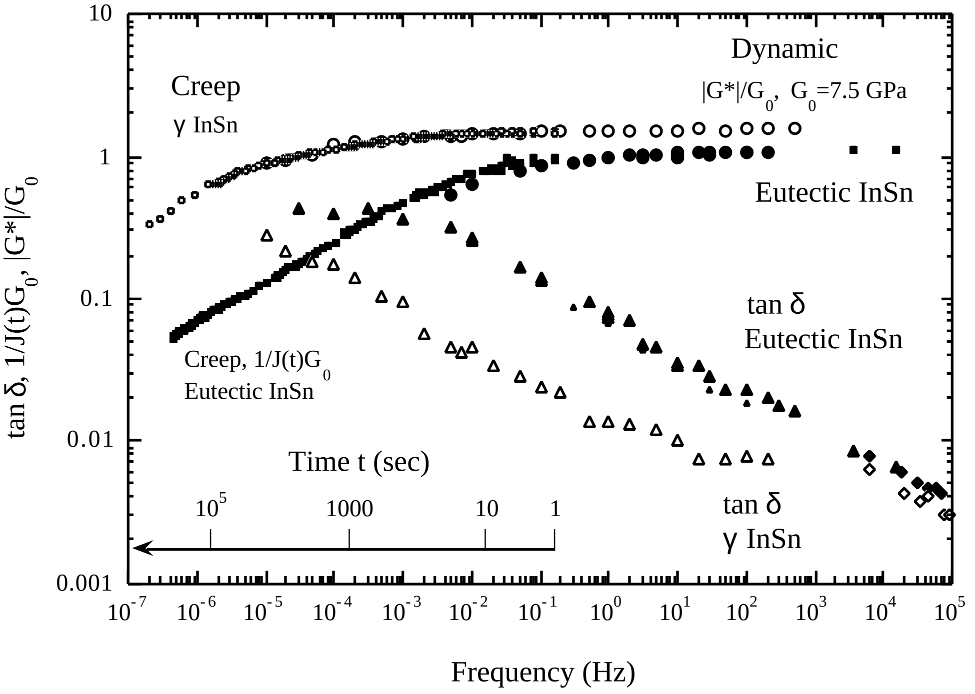


2. Viscoelastic behaviour of indium-tin at room temperature, both γ phase and eutectic. Δ, Δ: damping, tan δ; ◊, ◊, resonant damping. o, o: shear modulus |G*|. Inverse of the creep compliance as a function of time t and dynamic results as a function of frequency ν shown on the same plot via $2\pi\nu = t^{-1}$.


Creep
γ InSn
Dynamic
|G*|/G0, G0=7.5 GPa
Eutectic InSn
tan δ
Eutectic InSn
Creep, 1/J(t)G0
Eutectic InSn
Time t (sec)
10^5
1000
10
1
tan δ
γ InSn
10
1
0.1
0.01
0.001
10^-7
10^-6
10^-5
10^-4
10^-3
10^-2
10^-1
10^0
10^1
10^2
10^3
10^4
10^5
Frequency (Hz)
tan δ, 1/J(t)G0, |G*|/G0