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A relationship between non-exponential stress relaxation and delayed elasticity in the viscoelastic process in amorphous solids: Illustration on a chalcogenide glass



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ABSTRACT

Inorganic glasses are viscoelastic materials since they exhibit, below as well as above their glass transition temperature, a viscoelastic deformation under stress, which can be decomposed into a sum of an elastic part, an inelastic (or viscous) part and a delayed elastic part. The delayed elastic part is responsible for the non-linear primary creep stage observed during creep tests. During a stress relaxation test, the strain, imposed, is initially fully elastic, but is transformed, as the stress relaxes, into an inelastic and a delayed elastic strains. For linear viscoelastic materials, if the stress relaxation function can be fitted by a stretched exponential function, the evolution of each part of the strain can be predicted using the Boltzmann superposition principle. We develop here the equations of these evolutions, and we illustrate their accuracy by comparing them with experimental evolutions measured on GeSe₃ glass fibers. We illustrate also, by simple equations, the relationship between any kind of relaxation function based on additive contribution of different relaxation processes and the delayed elastic contribution to stress relaxation: the delayed elasticity is directly correlated to the dispersion of relaxations times of the processes involved during relaxation.

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1. Introduction

Many models (please see Phillips (1996) and references therein) have been proposed to explain the non-exponential relaxation in amorphous solids, and to give an “unified” theory, valid for any kind of relaxation, including stress relaxation. Nevertheless, the “delayed elasticity”, that inevitably emerges when the stress relaxation is not exponential, is not often discussed (see Goldstein (1969) where this problem is detailed) despite its preponderance during the first stages of relaxation. It is known, since more than a

century and a half (Weber, 1835), that the primary or transient creep stage of almost all kind of materials is due to delayed elasticity. It is not only due to a possible non-equilibrium viscosity, as astonishingly suggested by the ASTM standard for viscosity measurements (ASTM C1350-96, 1996), since glasses under equilibrium also exhibit primary creep stage. Delayed elasticity is also named “anelasticity” (mainly for metals (Zener, 1948)), or “retarded elasticity” (Goldstein, 1969) and was originally named “elastic aftereffect” (“*elastische nachwirkung*” (Boltzmann, 1876)). The delayed elastic deformation is a reversible deformation (“elastic”) which does not recover instantaneously (“delayed” or “retarded”) when the stress

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is released. The history of the investigation of the “aftereffect”, taking its origins in Göttingen with Weber and Kohlrausch, has been nicely summarized by Bendler (1984). The primary creep stage is supposed to occur in any kind of material, including ceramics, crystalline metallic materials, polymers, inorganic glasses, metallic glasses, as well as biomaterials. Thus, any kind of material is supposed to exhibit, during creep flow, delayed elasticity. Since nowadays a lot of commercial machines are available for measuring the steady state viscosity from the creep rate, it is sometimes forgotten that the creep rate is not solely controlled by the viscosity. During the primary creep stage, the creep rate is mainly controlled by the delayed elasticity, and the viscosity parameter can be measured only when the stationary creep stage is reached. This, for inorganic glasses, below their glass transition temperature, can take hours to days. Consequently, a “continuous viscosity measurement”, in the glass transition range, below or just above (down to a viscosity of, let say, 10^{9-10} Pa s), based on the continuous creep rate measurement, using continuous heating or cooling (no matter how slow it is), does not even make sense, and will obviously tend to give an “anomalous” apparent viscosity. In other words, non-isothermal viscosity measurements are necessarily wrong, because of the delayed elasticity. It underlines the necessity of a better knowledge of this delayed elastic deformation.

If it is quite straightforward to measure the delayed elasticity during a creep-recovery test, it becomes more delicate to evaluate it during a relaxation test. Wiechert (1893) is probably the first who has suggested that the delayed elasticity takes its origin in the non-exponentiality of the stress relaxation. A non-exponential relaxation can be modeled by a generalized Maxwell model (Maxwell, 1868) (or more precisely a “Maxwell–Wiechert model” (Wiechert, 1893)), where the relaxation modulus is decomposed into a Prony series. The equations, depicting the correlation between a non-exponential relaxation corresponding to a Maxwell–Wiechert model and the primary creep stage (including the delayed elasticity), have been set by Bennowitz and Rötger (see Simha (1942)). The Maxwell–Wiechert model is efficient to describe the viscoelastic behavior of silica glasses (Duffrène et al., 1997), but it requires a large number of parameters as compared to the well-known stretched exponential or “KWW” (Kohlrausch–Williams–Watts) function (Kohlrausch, 1854):

$$\varphi(t) = \frac{Q(t) - Q_\infty}{Q(0) - Q_\infty} = \exp\left(-\left(\frac{t}{\tau_0}\right)^\beta\right) \quad (1)$$

where φ is the relaxation function, describing the normalized relaxation of a quantity Q (here the shear stress), Q_∞ being its asymptotic value at $t \rightarrow +\infty$. β is the stretch exponent, and τ_0 is a characteristic time, related to the average (or “Maxwell’s”) relaxation time τ_a by the following relation:

$$\tau_a = \int_0^{+\infty} \varphi(t) dt = \frac{\Gamma(1/\beta)}{\beta} \tau_0 \quad (2)$$

where Γ is the Gamma function: $\Gamma(x, y) = \int_y^\infty s^{x-1} e^{-s} ds$ and $\Gamma(x) = \Gamma(x, 0)$. The average relaxation time corresponds to the average lifetime of an atom/molecule or group of

atoms/molecules in their equilibrium position (here when they are shifted by the stress) (Frenkel, 1926). The stretched exponential function (SEF) has various advantages. The main one is that it has only two parameters to describe the full relaxation spectrum. The second one is that the “departure” from the exponentiality is directly signed by the stretch exponent β . So, according to the idea of Wiechert, the delayed elasticity is directly signed by β . The SEF has been widely used to describe various kind of relaxation processes (see for example Phillips (1996)), but this function is not physically sound, first because regarding its time derivation:

$$\frac{d\varphi(t)}{dt} = -\varphi(t) \left(\frac{t}{\tau_0}\right)^\beta \beta t^{-1} \quad (3)$$

the relaxation rate tends to $-\infty$ when $t \rightarrow 0^+$, for $0 < \beta < 1$. Duffrène et al. (1997) have also pointed out the inadequacy of the SEF to describe the viscoelastic behavior of soda–lime–silica glasses. Additionally, they have shown that if we assume that the shear relaxation function corresponds to a SEF, the primary creep stage can not be fitted by another SEF, as it is often done.

In spite of the weaknesses of the SEF, we will show here, experimentally, how much the idea of Wiechert is correct: the delayed elasticity is only connected to the stretch exponent. The SEF is largely used in the literature mainly in order to describe the relaxation kinetic, but what is not often considered is its ability to describe the detailed mechanisms of relaxation, such as, for stress relaxation, the delayed elasticity and the inelasticity. We will illustrate, through relaxation-recovery tests on a viscoelastic material, how much efficient is the SEF to predict the evolution of the delayed elasticity. Then, we will show how we can interpret the relationship between any kind of relaxation function decomposable into a sum of exponential function (such as the SEF) and the delayed elasticity.

2. Theory

2.1. Linear viscoelasticity

Let us consider a linear viscoelastic body, undergoing a shear distortion, evolving over time: $\gamma(t)$, in the framework of the small strain assumption. According to the Boltzmann superposition principle (Boltzmann, 1876), the resulting shear stress is σ :

$$\sigma(t) = \int_{-\infty}^t G(t-s) \frac{d\gamma(s)}{ds} ds \quad (4)$$

G is the shear relaxation modulus of the viscoelastic body. Of course, considering the distortion as something imposed is just a point of view, and consequently, if we consider instead the stress as imposed, the resulting shear distortion is:

$$\gamma(t) = \int_{-\infty}^t J(t-s) \frac{d\sigma(s)}{ds} ds \quad (5)$$

J is the shear creep compliance. J and G are correlated by a Duhamel convolution equation (Ferry, 1980):

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