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A New Apparatus for Measuring High Viscosity of Solids[1]

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A new apparatus which is capable of measuring Newtonian viscosity as high as 10^8–10^{14} Pa·s has been developed. The upper and the lower faces of a cubic specimen $1 \times 1 \times 1$ cm^3 in size are respectively bonded to movable and fixed plates, and a constant shear stress is applied to the specimen. The displacement of the upper plate is measured and recorded as a function of time utilizing the HP-5528A optical laser measuring system. The sensitivity of the displacement measurement is 10 nm. Measurements at temperatures up to 200°C can be made by using a small furnace installed around the specimen. A variety of adhesive materials, polymers and ceramics, have been tested for the specimen–plate bonding. Aqueous Sauereisen cement showed a stable and rigid bonding in the experimental temperature range. Experiments for rubber (soft) and silicon crystal (hard) specimens have been performed to ascertain the reliability of the present method. The apparatus is also able to measure the anelasticity (mechanical relaxation) of solids, but is not adaptable to the measurement of the shear modulus of specimens; because the instantaneous displacement of the movable plate at the moment of the loading is mostly due to the rotational and inhomogeneous elastic deformations of the specimen.

KEY WORDS: anelasticity; laser interferometry; viscosity.

1. INTRODUCTION

Various materials generally exhibit viscoelastic behavior to some extent. For example, a solid under applied constant load gradually deforms with time. These dynamical properties are closely related to the microscopic character of materials, and the study of viscoelasticity is one of the many

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useful experimental methods in the solid-state physics of polymers, glasses, metals, composite materials, etc.

For the viscoelastic properties of materials, for example, Ferry [1] has reviewed experimental methods and analyses to derive theoretical constitutive equations. For measuring viscous properties of hard materials such as glasses, the beam-bending [2] and the fiber-elongation [3] methods have been frequently used. Here, the modes of deformation are far from a simple shear, which produces great complexities and difficulties in analyzing and interpreting experimental results. For measuring viscosities of soft metals (selenium), Cukierman and Uhlmann [4] have used the beam-bending method in the high-viscosity (low-temperature) range and the rotating cylinder viscometer in the low-viscosity (high-temperature) range. Carini et al. [5] have presented a new method for determining the high viscosity (up to 10^{14} Pa · s) of amorphous selenium using a holographic technique. In these experimental methods, the detection of small displacements of the specimen under the applied force is required. It should be noted that a laser interferometer can be used for measuring the displacement. For example, Imai and Bates [6] have determined the thermal expansion of solids using the above instrument. This method is also being used by us to study the anelastic and viscous properties of superionic conducting glasses such as AgI–$AgPO_3$ [7].

In this paper, we describe a new technique for measuring the viscoelasticity of Solids (viscosity, anelastic modulus, and anelstic relaxation time) by employing a laser interferometer and using a cubic test specimen. The use of a cubic specimen is for the purpose of experimental convenience. As shown later, one cannot obtain pure simple shear deformation of a solid by this method. However, the time-dependent deformation of the solid can be used to determine the dynamical shearing properties of viscoelastic materials.

2. APPARATUS

The principle of our method is illustrated in Fig. 1. A cubic specimen $1 \times 1 \times 1$ cm^3 in size is used, and the lower and upper faces of the specimen are bonded to fixed and movable stainless steel testing plates, respectively. A constant shearing load is applied to the movable plate by a weight and pulley. The lateral displacement of the plate is measured and recorded as a function of time by using the HP-5528A laser measurement system, which utilizes an optical heterodyne interferometric method. Two corner cube units, 1 cm in diameter, are used as shown in the figure, and they serve to reflect laser beams parallel to the incident beams. The nominal sensitivity of the displacement measurement is 10 nm. The temperature of the specimen

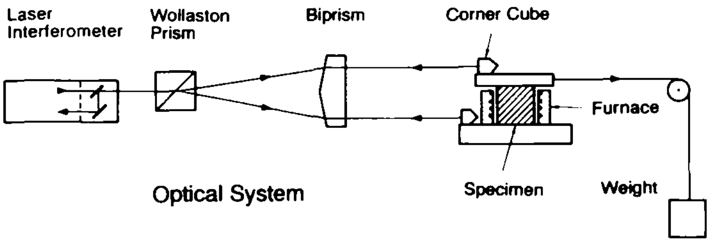


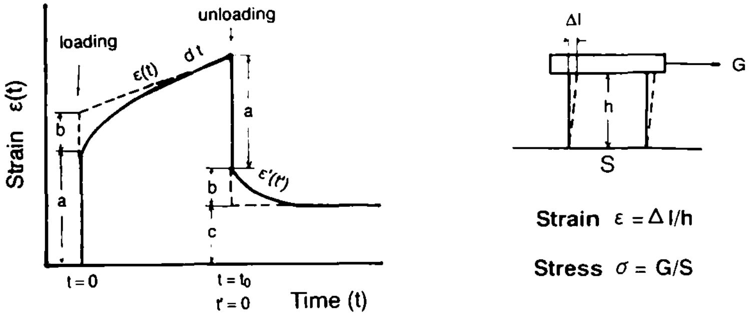
Fig. 1. Schematic diagrams of the apparatus.

is controlled by a small furnace, measured by a thermocouple attached to the upper face of the specimen, and controlled by a PID controlling unit. The constancy of the temperature was better than 0.1°C during a run of the experiment, which was 200–300 s in duration.

3. EXPERIMENTS

3.1. Analysis

The experimental data were analyzed as follows (see Fig. 2). For the moment, it is assumed that a simple shear stress mode is predominant for producing the viscoelastic deformation of the specimen. The propriety of this assumption will be reconsidered later. The load versus displacement relation is converted to the shear stress σ ($=G/S$; G is the load, S is the area to which the load is applied) versus shear strain ε ($=\Delta l/h$; Δl is the



$$\varepsilon(t) = a + b[1 - \exp(-t/\tau)] + d\,t$$

$$\varepsilon'(t') = c + b\exp(-t'/\tau)$$

Fig. 2. Deformation of the specimen.

displacement, h is the specimen height). When a constant stress σ is applied to the specimen at the time $t=0$ and then removed at $t=t_0$, the time-dependent strains $\varepsilon(t)$ and $\varepsilon'(t')$ are as shown in Fig. 2. When the anelasticity is due to a single mechanical relaxation, from standard anelasticity theory [8], the strains can be represented as follows:

$$t<t_0,\ \sigma\neq 0: \qquad \varepsilon(t)=a+b[1-\exp(-t|\tau)]+dt \tag{1}$$

$$t>t_0,\ \sigma=0: \qquad \varepsilon'(t')=c+b\exp(-t'/\tau) \tag{2}$$

where a, b, c, and d are parameters, and τ is the anelastic relaxation time.

These parameter values are used for determining the viscoelasticity. Concerning anelasticity, the relaxation time and the anelastic modulus are the most important quantities. Here, the anelastic modulus M_a, which is defined as the ratio between the applied stress and the anelastic strain, is used. Therefore, a small anelastic modulus corresponds to a strong anelasticity. By using the experimental data, the anelastic modulus can be calculated from the applied stress σ and the strain $\varepsilon_a=\varepsilon'(0)-\varepsilon'(\infty)=b$, namely $M_a=\sigma/b$. The viscosity η is, as usual, simply determined by definition as $\eta=\sigma/(d\varepsilon/dt)=\sigma/d$. It is noted here that in viscoelastic deformation, the strain $\varepsilon'(t')$ approaches the remaining strain produced by the viscous flow during the stressed state, namely $\varepsilon'(\infty)=c=dt_0$. Therefore, the visocity is also given as $\eta=\sigma/(c/t_0)$.

3.2. Rubber

In order to examine the adaptability of the assumption that the shear stress mode is predominant in producing the viscoelastic deformation of the specimen in the present experimental arrangement, the following test was performed. A rubber (Neoprene) cubic specimen of the usual size was used, and a 1-mm grid was printed on a side face of the specimen. The deformation of the specimen was visualized by taking a photograph of the side face at a magnification of about 20 times. In an example, the applied load was $G=308$ g and the displacement of the upper face was $\Delta l=1250\,\mu$m. The uniformity or deviation from the uniformity of deformation is scaled as follows. On the photographic picture, the (X, Y) coordinates of the lattice points were measured, where the origin of the coordinates was selected at the bottom-left lattice point. The differences of neighboring coordinate values, ΔX or ΔY, were plotted as a function of the position X $(=0\text{–}10)$ with the position Y as a parameter as shown in Fig. 3. Here, the quantities ΔX and ΔY were normalized by their values near the central lattice point. The values of ΔX and ΔY for $2<Y<9$ were not very different from those for $Y=5$, and are not shown in the figure. The scatter

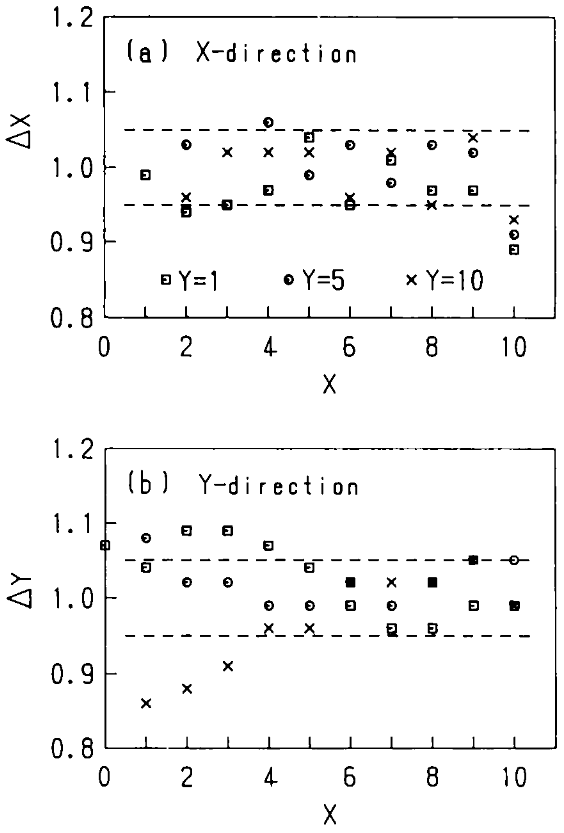


Fig. 3. Strain distributions in the rubber specimen.

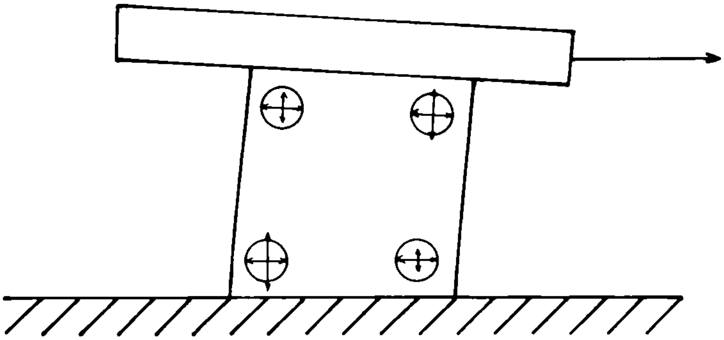


Fig. 4. Pattern of the deformed rubber specimen.

of the ΔX or ΔY values within the range of $0.95 < \Delta X, \Delta Y < 1.05$ may be due to measurements errors and may not be meaningful. In Fig. 3a, a slight decrease of ΔX (extra compression along the X direction) is only observed near $X = 10$. In Fig. 3b, an increase and a decrease of ΔY (extra extension and extra compression along the Y direction) are seen for $Y = 1$ and $Y = 10$. These kinds of nonuniformities show deviations from the simple shear mode. However, it is seen that the nonuniformity along the X direction is slight and very restricted spatially, while the nonuniformity along the Y direction is not so small. These situations are illustrated in Fig. 4, showing an S-shaped deformation. Here, the arrows indicate the displacements of grids and the circles show their uniform displacements.

As a result, we conclude that the deformation in the X direction is almost pure simple shear, although the deformation is not simple shear in the Y direction. Note that the viscoelastic behavior of specimens is observed along the X direction, and the above situation is acceptable for the purpose of our experiment.

In order to confirm the reliability of this method, we measured the viscosity of the superionic conducting glass $(AgI)_{0.5}(AgPO_3)_{0.5}$ [9] using a rotational method in the range of 10^{-2}–10^3 Pa·s, an ocillational method for 10^3–10^8 Pa·s, and the present method for 10^8–10^{14} Pa·s. These data presented a smooth curve in a whole range of the measured viscosities.

3.3. Silicon

A high-purity silicon single crystal was used as a test specimen and the results are shown in Fig. 5. Silicon was rigid and was scarcely deformed at low temperatures under the small applied load in the present experiment. Therefore, the observed displacement is considered wholly due to the bonding material used between silicon and the testing plates. In the present study, various kinds of polymer adhesives and ceramic cements were tested: Aron-alpha, Araldite, Polymid, Sauereisen, Ceramabond, and Sumiceram. The most favorable one was, even at elevated temperature, Sauereisen No. P-1 (Sauereisen Cement Co., Pittsburgh, PA). As seen in Fig. 5a, a large instantaneous deformation and flow deformation were observed in the case of Aron-alpha when the temperature T was somewhat elevated. In contrast, Sauereisen cement seemed to be stable even at higher temperatures, as seen in Fig. 5b. In this case, a very slight displacement occurred when the load was applied, and this disappeared when the load was removed. This may be due to a small elastic deformation of the bonding material. The purpose of our study is to determine the viscoelastic properties of specimen materials, and the absence of flow deformation of adhesives, not an elastic one, is necessary. Thus, Sauereisen cement was usually utilized in our experiments.

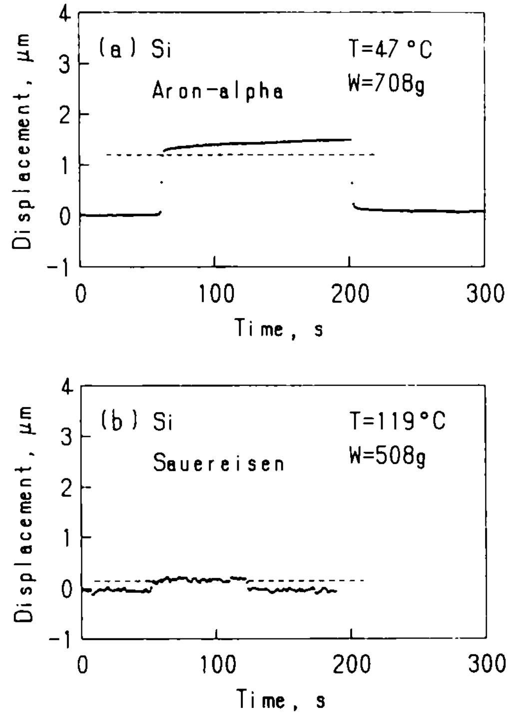


Fig. 5. Displacement versus time for Si with (a) polymer and (b) cement adhesives.

4. CONCLUSIONS

The characteristics of our experimental method are summarized as follows.

(a) The viscoelastic behavior (anelasticity and viscosity) of solids can be measured accurately owing to the highly sensitive optical method.

(b) The stresses activating the viscoelastic deformation are almost of the simple shear type, which makes it possible to simplify the experimental situation. This is very different from the situation in other viscosity-measuring methods such as the beam-bending method.

There are, however, some restrictions with this method and also some problems to be carefully considered.

(c) The most important problem is finding an adequate adhesive to fix the specimen to the testing plates. The bond should be stable enough to contribute no appreciable flow deformation and rigid enough to be durable to applied loads for desired ranges of stress and temperature.

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