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Pressure Dependence of the Creep of Lead*

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Apparatus is described for maintaining hydrostatic environment, 1 in. in diameter by 4 in. long, with 8 to 12 electrical leads, up to 20-kbar pressure and various temperatures. Bending creep of 99.999+% lead is reported with an activation volume of about 21×10^{-24} cm^3, essentially independent of temperature between 0° and 57°C. Evidence for recrystallization is given.

INTRODUCTION

THE effect of pressure on properties of materials has been extensively reviewed.[1–3] Theories of creep have also been reviewed elsewhere.[4–6] Those relating creep to self-diffusion involve dislocation climb,[7,8] vacancy creep,[9,10] and motion of "joggy" dislocations[11] among other mechanisms. Bridgman[12] has investigated the effect of pressure on Young's modulus, the shear modulus, work hardening, elastic limit, ultimate strength, etc., for a large number of materials. Lazarus,[13] Daniels and Smith,[14] and Hughes *et al.*[15] have studied the effect of pressure on elastic constants of a number of solids. Christy[16] has reported the effect of pressure on creep in silver bromide. Butcher and Ruoff[17] have investigated the effect of pressure in 99.999% lead. The influence of pressure on self diffusion in sodium,[18] phosphorus,[19] zinc,[20] silver,[21] silver bromide,[22] and lead[23] has been found to approximate an equation of the type

$$D=D_0e^{-p\Delta V^\ddagger/KT}, \tag{1}$$

where D is the diffusion coefficient, D_0 the value of D at zero pressure, p the pressure, $\Delta V^\ddagger$ the activation volume, T the temperature, and K Boltzmann's constant. Nachtrieb, Resing, and Rice[23] found a temperature-independent activation volume of 0.71 to 0.87 atomic volumes for self-diffusion in 99.999% lead between 253° to 301°C.

If creep were vacancy controlled, the activation volume for creep would be expected to be equal to that for self-diffusion, and hence given by

$$\Delta V^\ddagger=[KT/(p_2-p_1)]\ln(\dot\epsilon_1/\dot\epsilon_2), \tag{2}$$

where $\dot\epsilon_1$ is the deformation rate at pressure p_1. Butcher and Ruoff[23] reported the activation volume for creep in lead to be 0.80 atomic volumes at 70°C.

SAMPLE PREPARATION

Samples were prepared from 99.999+% lead.[24] Typical principal impurities were listed as Ag, Cu, Fe, and Bi, less than 1 ppm each. An ingot of the material was crushed in a press until it was approximately 0.375 cm thick. Beams were carefully cut from this with a hacksaw and carefully filed or sanded to various sizes 0.28 to 0.38 cm thick by 0.3 to 0.4 cm wide and 2.4 cm long, the sizes chosen to obtain about the same creep rate at different temperatures. Visual inspection of surfaces of the samples, after finishing revealed that the grain size varied from less than 0.1-mm to 1-mm diameter, with a mean size of approximately 0.5 mm. Sample 20 was melted and allowed to cool slowly, resulting in the center $\frac{2}{3}$ of the sample's being one large grain.

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APPARATUS AND EXPERIMENTAL PROCEDURE

The composite high-pressure vessel used in these studies is shown schematically in Fig. 1. With piston and bottom plug in place, it provided a working space 1 in. in diameter by 4 to 5 in. long. It has been tested to 20 kbar, although the present creep measurements

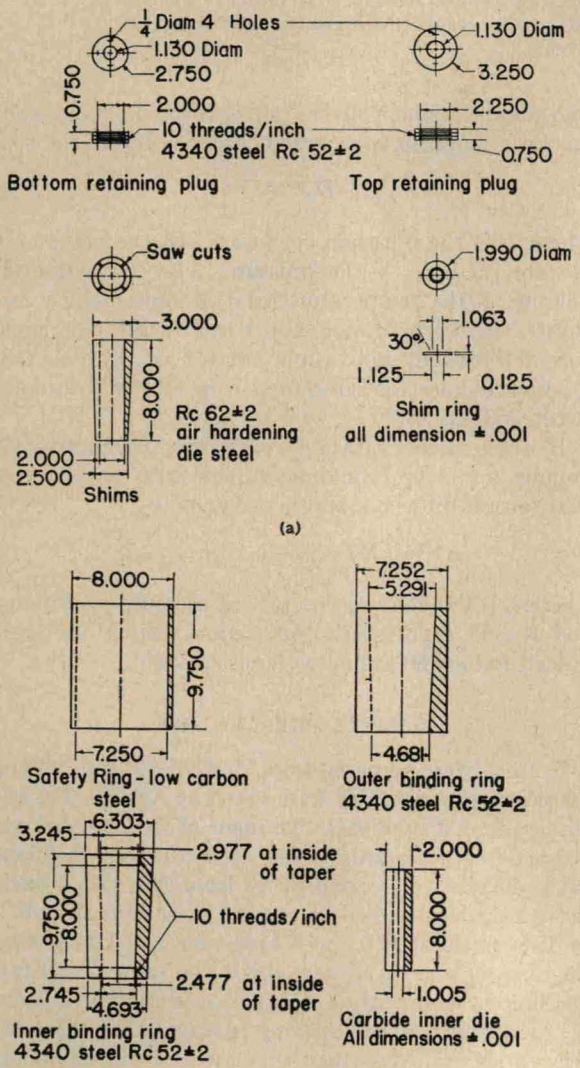

FIG. 1. (a) and (b), high-pressure vessel component parts shown in partial cross section. Safety ring, outer binding ring, inner binding ring, shims, and die are assembled successively in a hydraulic press. Shim rings are then introduced between both screw-in retaining rings and die.

were only carried on up to 10 kbar. The piston was sealed with a Bridgman unsupported area seal, using polyethylene as the soft seal material. The bottom plug, shown in Fig. 2, introduced 8 (or sometimes 12) electrical leads into the pressure vessel. Figure 2 also illustrates the means by which the creep apparatus was mounted on the bottom plug for easy assembly

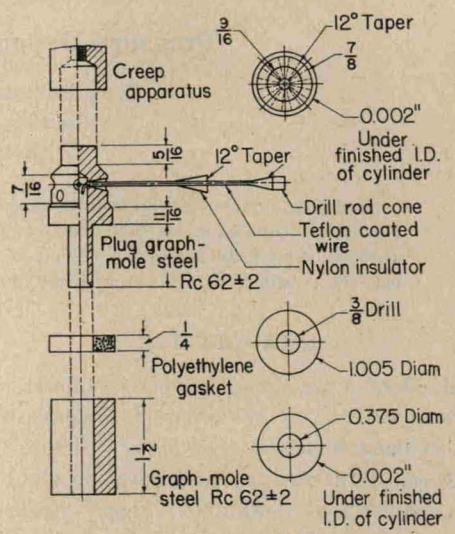

FIG. 2. High-pressure plug with 8 electrical leads (dimensions are in inches).

in the pressure vessel. Pressure was determined from the force applied to the piston, calibrated with 2.1% gold-chrome[3] and also manganin[1,3] coils.

The temperature was controlled by a continually stirred water bath in which the pressure vessel was completely immersed. Heat was provided by a 1 kW immersable heater regulated by a mercury control and relay.[25] The temperature varied by less than $\pm\frac{1}{2}$°C as continually recorded on a Bristol Dynamaster dc potentiometer. In room-temperature studies, the ambient was controlled to ±2°C. Low-temperature studies were made with the bath packed in ice, with a maximum temperature variation of 2°C during creep.

Figure 3 shows schematically the creep stressing apparatus, which was surrounded by a stainless steel tube. The sample rested in slots in the tube which served as knife edges for 3-point loading. The other knife edge consisted of a wire loop attached rigidly to the spring which supplied the stressing force. Since a spring was used to load the sample, the load changed slightly with deformation. However, weak springs with

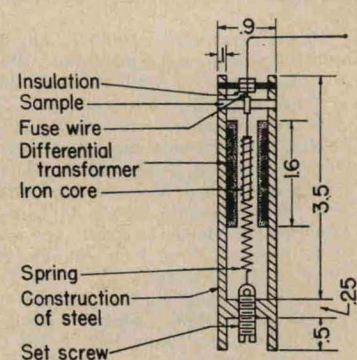

FIG. 3. Cross section of creep apparatus (dimensions are in inches).

[25] Type CF-708, Philadelphia Scientific Glass Company, Quakertown, Pennsylvania.

high elongation were used to keep this change in force to 5% or less. Pressure has an effect on the spring constant through its effect on the shear modulus.[26] This effect in steel is of the order of a percent or two at the pressures used here and so has been neglected. A fuse wire was employed to hold the load off the sample until the desired temperature and pressure were obtained, at which time the creep was commenced by fusing the wire.[27]

The sample deflection was measured by a differential transformer wound on a polyethylene core. The primary coil was composed of 600 turns of 38 gauge copper magnet wire. Each of the secondaries consisted of 3000 turns of the same wire. The entire assembly was approximately 4 cm in length, 1.75-cm outside diameter, and 1-cm inside diameter. It was mounted in the stressing apparatus as shown in Fig. 3. The core was soft iron, slightly smaller than the inside diameter of the coil spring. The surface of the core was carefully polished and the end rounded to avoid interference with motion of the spring. This core was rigidly attached to the upper end of the spring and center knife edge.

The primary coil was driven at 1000 to 2000 cps by a Hewlett Packard 200T precision telemeter test oscillator. The signals from the secondary coils were demodulated and their difference recorded on a Leeds and Northrup 10-mV Speedomax recorder, modified to read 10 mV, 100 mV, etc. up to 300-V full scale. Most of these studies were made on the 100-mV scale. Calibration with a soft iron core mounted on a micrometer, and visual inspection of a deforming sample showed the differential transformer output to be linear with core displacement for a region slightly greater than 1.25 cm in length. The output was approximately 5 V per cm which resulted in a sensitivity with the present measuring system of at least 5×10^{-5} cm. A number of the experiments were made with the driving voltage lowered to give a sensitivity of 1 V per cm to facilitate recording over very long times.

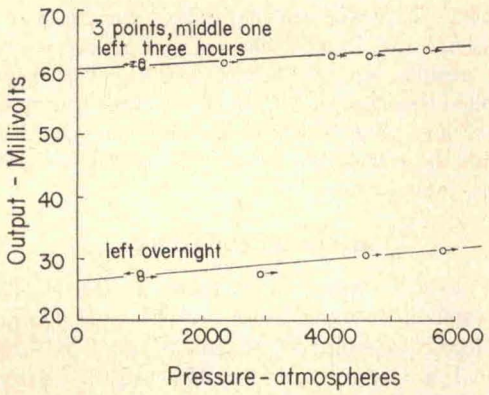

FIG. 4. Pressure calibration of displacement gauge. Lower curve shows pressure dependence of output using porcelain sample, the upper curve indicates output with knife edge rigidly held above the beam. A pressure dependence of displacement reading would cause a deviation from parallelism.

To determine the effect of pressure on the voltage–displacement characteristics of the differential transformer, the core was securely fixed in one position with the fuse wire, and the lead sample was replaced by a porcelain beam. With the core so fixed, the pressure was varied and the effect on output noted. The fuse wire was then broken, allowing the core to drop a few millimeters, being supported in this position by the porcelain beam. The results are shown in Fig. 4. It can be noted that the curves are nearly parallel, indicating a very small effect of pressure on the displacement reading. The voltage output for the same displacement was ~3% higher at 6 kbars than at atmospheric pressure. The stability of the transducer readings is indicated by the amount of drift occurring overnight and in 3 h, respectively.

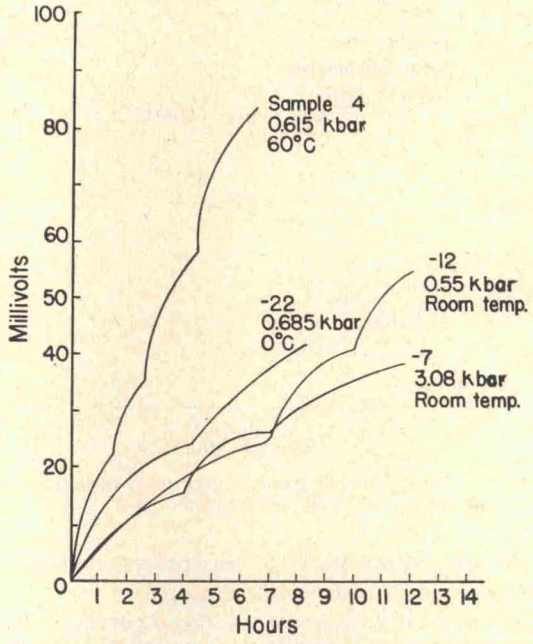

FIG. 5. Typical creep curves at various pressures and temperatures. (Sensitivity 1 V/cm this figure only.)

RESULTS AND DISCUSSION

Figure 5 shows a number of creep curves taken at various temperatures and pressures. Creep deflection is given in mV output. Figure 6 shows a typical creep curve illustrating the effect of sudden pressure changes.

The curves in Fig. 5 are not smooth, but rather have regions of acceleration that give them a wavy appearance, suggestive of recrystallization.[28] The notion of

[26] P. W. Bridgman, Proc. Am. Acad. Arts Sci. **64**, 39 (1929).

[27] For a time the experiments were harrassed by transversal fractures of the inner die of the pressure vessel, initiated when a free end of the 12-V fuse wire shorted out there.

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recrystallization was strengthened by the behavior of sample 20. After creep at room temperature and 5 kbar, similarly to the other samples, visual inspection of the surface showed the average grain size had been reduced to approximately 0.5 mm in the formerly monocrystalline region. Since recrystallization had occurred with such a large grain it is likely that recrystallization also took place in other samples, since finer-grained material, in general, recrystallizes more readily.[6] The waves in the creep curve might be explained as follows: At various stages of creep, different points in the sample beam may reach strains sufficient to initiate recrystallization at the temperature and pressure of the experiment. The recrystallization might remove work hardening present in that region of the sample, resulting in an increase of the deformation rate. As

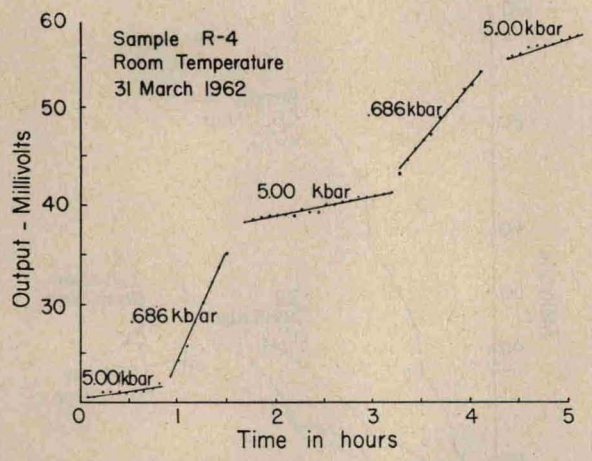

FIG. 6. Typical room temperature creep curve at various pressures (dotted curve). Solid line indicates "best" slope.

macroscopic strain proceeds, this process might be repeated at various places on the sample.

The wavy appearance of the creep curves made interpretation of data similar to that of Fig. 6 difficult and gave rise to quite a bit of scatter. A "best" slope from each of the intervals of Fig. 6 was determined, and these slopes plotted logarithmically against pressure for each sample. Studies were carried out at pressures up to 10 kbars. Equation (2) was used in the determination of the activation volume. In the use of this equation the creep rate in one pressure interval was compared with the average of the pressure intervals before and after it. This may tend to compensate for the change in stress with sample deflection. Figure 7 shows the activation volumes deduced for creep as a

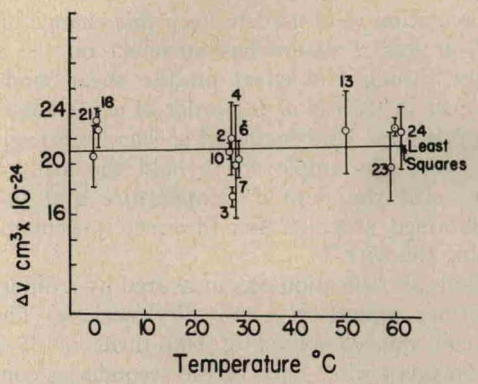

FIG. 7. Temperature dependence of activation volume. Figures indicated samples. Circles indicate average values for a given sample, error bars denote extreme values. Horizontal line represents least squares fit of all measurements.

function of temperature. The center point in each case was taken as the geometric means of the points for any given sample. The uncertainty brackets indicate maximum deviation from these means. The points of maximum deviation from the mean always occurred either early in the run where rather rapid work hardening was taking place, or very late on the creep curve where the curvature of the beam was large. The least-squares average of all the points is approximately 21×10^{-24} cm^3 or about $\frac{2}{3}$ of the room temperature, room pressure atomic volume of $\sim30.3\times10^{-24}$. If only the "well behaved" portions of each creep segment were used in the activation volume calculations, all points could be satisfied by $\Delta V^{\ddagger}=21\pm4\times10^{-24}$ cm^3. Within the accuracy of these studies no change in activation volume with temperature was indicated.

Within experimental error the activation volume for creep determined in these experiments agrees with that for self-diffusion as determined by Nachtrieb, Resing, and Rice[23] at 253°C, and with those for creep at 70°C by Butcher and Ruoff[17] who claim a greater accuracy. These results are then compatible with diffusion-controlled theories, such as vacancy climb and dragging along of jogs. The Nabarro–Herring[9,10] mechanism can probably be eliminated because of rate considerations at these temperatures.

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