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Elastic properties of Pd$_{40}$Cu$_{30}$Ni$_{10}$P$_{20}$ bulk glass in supercooled liquid region

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In situ ultrasonic measurements for the Pd$_{40}$Cu$_{30}$Ni$_{10}$P$_{20}$ bulk glass in three states: Glassy solid, supercooled liquid, and crystalline, have been performed. It is found that velocities of both longitudinal and transverse waves and elastic moduli (shear modulus, bulk modulus, Young’s modulus, and Lamé parameter), together with Debye temperature, gradually decrease with increasing temperature through the glass transition temperature as the Poisson’s ratio increases. The behavior of the velocity of transverse wave vs. temperature in the supercooled liquid region could be explained by viscosity flow, rather than the two different crystallization processes in the region, suggested in the literature. No decomposition was detected at a temperature only 5 K below the crystallization temperature. © 2001 American Institute of Physics. [DOI: 10.1063/1.1359490]
slope of 1.890 m/(sK). However, between \( T_g \) and \( T_x \), \( V_s \) rapidly drops from about 1.792 km/s at 580 K to 1.719 km/s at 614 K. It has a constant value of roughly 1.712 km/s around 630 K, followed by a rapid increase to 1.771 km/s at 635 K. Above \( T_x \), the temperature dependence of both waves is complex. The sample first consists of crystalline phases and residual amorphous alloy, and then crystallizes completely above about 660 K. Using the density data of Pd\(_{40}\)Cu\(_{30}\)Ni\(_{10}\)P\(_{20}\) bulk glass versus temperature reported in Ref. 8, the temperature dependences of the elastic moduli (shear modulus, Young’s modulus, Lamé parameter, and bulk modulus) together with Debye temperature and Poisson’s ratio, can be found in Figs. 3 and 4. At 300 K, the shear modulus, Young’s modulus, Lamé parameter, bulk modulus, Debye temperature, and Poisson’s ratio are 33.4, 93, 123, 145.3 GPa, 270 K, and 0.393, respectively. These values are similar to those for Pd\(_{39}\)Cu\(_{30}\)Ni\(_{10}\)P\(_{21}\) bulk glass. \(^{12}\) With increasing temperature the shear modulus, Young’s modulus, Lamé parameter, bulk modulus, and Debye temperature decrease, while the Poisson’s ratio increases. At \( T_g \), there is no dramatic change of any of these parameters, although the slope of specific volume vs temperature changes from \( 5.457 \times 10^{-3} \) m\(^3\)/K below \( T_g \) to \( 8.785 \times 10^{-3} \) m\(^3\)/K above \( T_g \). \(^{8}\) Note that a significant change in shear modulus at \( T_g \) was suggested for ZrTiCuNiBe bulk glasses. \(^{16}\) In the supercooled liquid region, it is experimentally found that the temperature dependences of all the elastic moduli, the Debye temperature, and Poisson’s ratio, are strongly correlated with the temperature dependence of the velocity of transverse waves. The Debye temperature drops from about 252 K at 600 K to 243 K at 625 K, indicating that interatomic interactions become weaker. This is also reflected in the shear modulus and Young’s modulus. On the other hand, Poisson’s ratio increases from 0.402 to 0.406, while bulk modulus and Lamé parameter have a maximum around 610 K. At the crystallization temperature, \( T_x \), a kink is detected for all elastic constants. Above \( T_x \), the temperature dependence of all elastic constant is complicated due to the sample microstructure.

To explain the behavior of the velocity of transverse wave vs. temperature in the supercooled liquid region, microstructures of samples annealed at 615, 630, and 650 K were investigated by high-resolution transmission electronic microscopy (HRTEM). Some examples are shown in Fig. 5. The samples were heated in the DSC at a rate of 5 K/min to the given temperature. After annealing for less than 5 s, the samples were then quenched to room temperature. It was found that after annealing at 615 and 630 K respectively, the materials were still in the amorphous state. No decomposition was detected in samples annealed at 615 and 630 K, while the material partially crystallized after the annealing at 650 K. Longitudinal waves are readily propagated in gases and liquids, as well as in elastic solids. The gradual decrease of the velocity of longitudinal waves with temperature in Fig. 2 is a result from the softening of the interatomic inter-

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**FIG. 2.** Velocities of longitudinal (\( V_l \)) and transverse waves (\( V_s \)) vs. temperature for \( \text{Pd}_{40}\text{Cu}_{30}\text{Ni}_{10}\text{P}_{20} \) bulk glass at a heating rate of 5 K/min under a flow of purified Ar. \( T_g \) and \( T_x \) are indicated.

**FIG. 3.** Temperature dependences of the elastic moduli [shear modulus (G), bulk modulus (K), Young’s modulus (E)] and Lamé parameter (\( \lambda \)), for \( \text{Pd}_{40}\text{Cu}_{30}\text{Ni}_{10}\text{P}_{20} \) bulk glass. \( T_g \) and \( T_x \) are indicated.

**FIG. 4.** Temperature dependences of the Debye temperature (\( \theta_D \)) and Poisson’s ratio for \( \text{Pd}_{40}\text{Cu}_{30}\text{Ni}_{10}\text{P}_{20} \) bulk glass. \( T_g \) and \( T_x \) are indicated.

**FIG. 5.** High-resolution transmission electron microscopy images and their corresponding selected area diffraction patterns of samples heated at a rate of 5 K/min to 615, 630, and 650 K, respectively, and annealed for less than 5 s before being quenched to room temperature.
action. No kink was observed at $T_x$ and $T_g$, while $V_t$ has a minimum at about 658 K where the sample is completely crystallized. Unlike longitudinal waves, transverse waves are not supported by elastic collisions of adjacent atoms. For the propagation of transverse waves, it is necessary that each atom attracts its neighbors strongly so that during movement it pulls its neighbors with it, thus causing the sound to move through the material. Consequently, in gases, the forces of attraction between molecules are too small for shear waves being transmitted. The same is true for liquids, unless it is the longitudinal and the transverse wave velocity in the supercooled liquid region from 580 to 620 K, and the crystallization reaction at 635 K, could explain the observed behavior of transverse wave velocity vs. temperature in the 580–635 K region.

\[ V_{\text{t}} \approx 10^{12} \text{ to } 10^8 \text{ Pa s} \text{ in the supercooled liquid region}, \]

\[ \text{rapidly increases when approaching the crystallization temperature.} \]

Thus, viscous flow in the supercooled liquid region from 580 to 620 K, and the crystallization reaction at 635 K, could explain the observed behavior of transverse wave velocity vs. temperature in the 580–635 K region.\[ \text{In situ} \]

ultrasonic measurements for bulk Pd$_{40}$Cu$_{30}$Ni$_{10}$P$_{20}$ glass in three states: glassy solid, supercooled liquid, and crystalline, have been carried out. From the results of viscosity versus temperature,\[ \text{it appears quite probable that the observed temperature dependency of both the longitudinal and the transverse wave velocity in the supercooled liquid region results mainly from viscous flow. No evidence of the two different crystallization processes suggested in the literature was observed in the supercooled liquid region, and there are no dramatic changes in the elastic moduli (the shear modulus, bulk modulus, Young’s modulus, and Lamé parameter), and in the Debye temperature and Poisson’s ratio at the glass transition temperature. Furthermore, no decomposition was detected in a sample annealed at a temperature just 5 K below the crystallization temperature.} \]

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