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Evaluation of the elastic properties of bulk metallic glasses

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Abstract The development of bulk metallic glasses as a prominent class of functional and structural materials has attracted considerable interest in the last years. One of the fundamental physical quantities necessary to describe the mechanical properties of the materials is the bulk modulus. In the present article, a simple method to estimate the bulk modulus and its pressure derivative is proposed. It is shown that these quantities can be estimated from the values of the constituent elements and their compositions. Comparison with measured data shows good agreement. The physical background of the method is discussed based on the jellium model of metals.

Keywords Bulk metallic glasses · Bulk modulus · Jellium model · Pressure

Introduction

During the last decade, new multicomponent bulk amorphous alloys called bulk metallic glasses have been developed and studied intensively [1–7]. In contrast to the traditional amorphous metals, these new alloys have low critical cooling rates and fully amorphous samples with one side dimension as large as 1 cm can be produced by conventional processes. The bulk metallic glasses are characterized by the high thermal stability of their supercooled liquids, which permit the study of thermophysical properties in the supercooled liquid in addition to the amorphous solids [8, 9]. Bulk metallic glasses have many unique properties such as extra ordinary high strength, low ductility, high hardness, and excellent corrosion resistance.

One of the fundamental physical quantities necessary to describe the mechanical properties of materials is the bulk modulus. For an efficient application of the physical properties of metallic glasses, a good understanding of this quantity is necessary. The development of theoretical methods to estimate the values of the bulk modulus is also necessary. The calculation of physical quantities through the use of traditional methods such as those based on electronic structure calculations could be accurate. However, it is technically too involved, time consuming and not appropriate to study the general trend of the materials in a simple way. Therefore, it will be valuable to develop a method to estimate easily the physical quantities. In the present contribution, it is shown that the bulk modulus and its pressure derivative of the bulk metallic glasses can be estimated from the values of the constituent elements and their compositions. By comparing the estimated values with the measured ones, we show that the method is predictive. The physical background of the method is discussed based on the jellium model of metals.

Estimation of bulk modulus and its pressure derivative

The simplest way to calculate the physical quantity Y of a complex system is

$$Y = \sum_i c_i Y_i, \qquad \sum_i c_i = 1, \tag{1}$$

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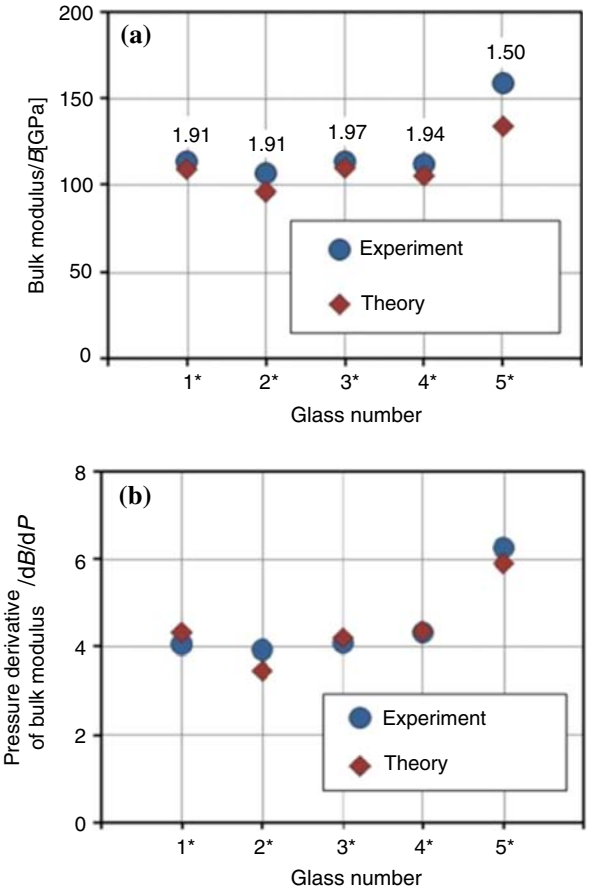

Fig. 1 Comparison between the calculated and the measured data of bulk modulus (**a**) and its pressure derivative (**b**). The numbers indicate the following glasses: (1) $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$, (2) $Zr_{41}Ti_{14}Cu_{12.5}Ni_9Be_{22.5}C_1$, (3) $Zr_{48}Nb_8Cu_{12}Fe_8Be_{24}$, (4) $(Zr_{0.59}Ti_{0.06}Cu_{0.22}Ni_{0.13})_{85.7}Al_{14.3}$, and (5) $Pd_{39}Ni_{10}Cu_{30}P_{21}$. The asterisks are used to avoid confusion with the glass number of Table 1. The numbers in (a) denote the values of the electron density parameter r_s in units of [au] = [0.53 × 10^{-10} m]

where Y_i is the physical quantity of the constituent element i and c_i is its concentration. Here, we calculate for the cases of bulk modulus B and its pressure derivative dB/dP. Values of B and dB/dP for the elemental systems have been taken from the literature [10, 11]. The comparison between the calculated and the measured data in bulk metallic glasses is shown in Fig. 1. We can see that the calculated results are in good agreement with the experimental data [12].

The result shown in Fig. 1 indicates that the method is effective for the estimation of B and dB/dP of bulk metallic glasses. Therefore, we can predict easily these values. The predicted values of B for some systems waiting for experimental verifications are shown in Fig. 2. The numbers in the figure indicate the glasses given in Table 1. In Fig. 3, the predicted values of dB/dP is shown.

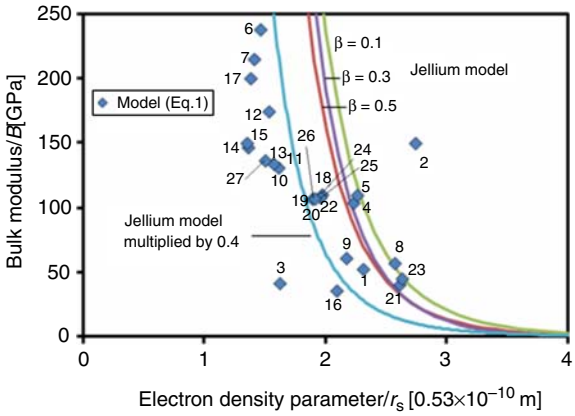

Fig. 2 The predicted values of the bulk modulus B (calculated by Eq. 1) are shown as a function of the electron density parameter r_s. The numbers in the figure indicate the glasses given in Table 1

Table 1 Glasses shown in Figs. 2, 3

No.	Glasses
1	$La_{55}Al_{25}Ni_5Cu_{10}Co_5$
2	$Au_{55}Cu_{25}Si_{20}$
3	$Ce_{70}Al_{10}Ni_{10}Cu_{10}$
4	$Cu_{46}Zr_{42}Al_7Y_5$
5	$Cu_{60}Zr_{20}Hf_{10}Ti_{10}$
6	$Fe_{53}Cr_{15}Mo_{14}Er_1C_{15}B_6$
7	$Fe_{61}Mn_{10}Cr_4Mo_6Er_1C_{15}B_6$
8	$Mg_{65}Cu_{25}Gd_{10}$
9	$Nd_{60}Al_{10}Fe_{20}Co_{10}$
10	$Ni_{40}Cu_5Ti_{17}Zr_{28}Al_{10}$
11	$Ni_{45}Ti_{20}Zr_{25}Al_{10}$
12	$Ni_{60}Nb_{35}Sn_5$
13	$Pd_{40}Cu_{30}Ni_{10}P_{20}$
14	$Pd_{60}Fe_{20}P_{20}$
15	$Pd_{64}Ni_{16}P_{20}$
16	$Pr_{60}Cu_{20}Ni_{10}Al_{10}$
17	$Pt_{60}Ni_{15}P_{25}$
18*	$Zr_{48}Nb_8Cu_{12}Fe_8Be_{24}$
19	$Zr_{55}Al_{19}Co_{19}Cu_7$
20	$Zr_{57.5}Cu_{15.4}Ni_{12}Al_{10}Nb_5$
21	$Mg_{70}Zn_{25}Cu_5$
22	$Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$
23	$Mg_{65}Cu_{25}Tb_{10}$
24*	$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$
25*	$Zr_{41}Ti_{14}Cu_{12.5}Ni_9Be_{22.5}C_1$
26*	$(Zr_{0.59}Ti_{0.06}Cu_{0.22}Ni_{0.13})_{85.7}Al_{14.3}$
27*	$Pd_{39}Ni_{10}Cu_{30}P_{21}$

Glasses considered in Fig. 1 are marked with asterisks

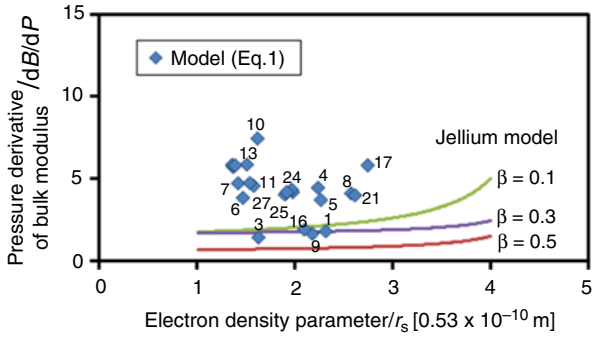

Fig. 3 The predicted values of the pressure derivative of the bulk modulus dB/dP (calculated by Eq. 1) are shown as a function of the electron density parameter r_s. The numbers in the figure indicate the glasses given in Table 1. To avoid congestion, some glass numbers are not shown

Discussion

The agreement between the predicted and the measured values of B and dB/dP shown in Fig. 1 is surprising, in view of the simplicity of the method of calculation. It should be mentioned here that Eq. 1 was also used to analyze the measured data of some bulk metallic glasses [12]. However, it has never been used to predict unmeasured quantities nor a physical interpretation has been given. The result given in Fig. 1 suggests that the essence of the metallic bonding operating in a complex real system such as bulk metallic glasses can be rationalized in terms of simple models. In this section, the probable physical background of the method is discussed based on the jellium model of metals.

Calculations based on jellium model indicate that the material trend in the bulk modulus is determined essentially by the electron density [13]. We have used this notion to interpret our result. According to the jellium model, the energy of the interacting electron gas that incorporates the kinetic, exchange, and correlation energies is written as [14, 15]

$$\varepsilon = \frac{3}{5\alpha^2 r_s^2} - \frac{3}{2\pi\alpha r_s}(1+\beta), \tag{2}$$

$$\alpha = \left(\frac{4}{9\pi}\right)^{1/3}, \tag{3}$$

$$\frac{1}{n} = \frac{4}{3}\pi r_s^3, \tag{4}$$

where β represents the contribution from the correlation energy and n is the electron number density. The correlation energy is always negative and at metallic densities, the value of β goes from 0.1 to 0.5 [15]. The bulk modulus and its pressure derivative are written as

$$B = \frac{1}{12\pi r_s}\left(\frac{\partial^2\varepsilon}{\partial r_s^2} - \frac{2}{r_s}\frac{\partial\varepsilon}{\partial r_s}\right), \tag{5}$$

$$\frac{\partial B}{\partial P} = -\frac{1}{3}\frac{r_s}{B}\frac{\partial B}{\partial r_s}, \tag{6}$$

$$\frac{\partial B}{\partial r_s} = -\frac{5}{2\pi\alpha^2}\frac{1}{r_s^6} + \frac{2}{\pi^2\alpha}(1+\beta)\frac{1}{r_s^5}. \tag{7}$$

The above equations indicate that the values of B and dB/dP are obtained if the electron number density parameter r_s is available. Concerning the values r_s for the elements, many researchers have reported their own values based on different degree of sophistication [16–19]. Although there are differences in the details, all the results show the same behavior and trend along the periodic table of the elements [19, 20]. Based on this observation, we have calculated the values of r_s by using the formula [21]

$$r_s = 1.388 a_B \left(\frac{A}{z\rho}\right)^{1/3} \tag{8}$$

where a_B is the Bohr radius. A, z, and ρ are the molecular mass, number of valence electrons, and mass density per formula unit, respectively. Values of ρ have been taken from [12, 22]. It should be remarked that the simple number of valence electrons counting method have been used successfully in the study of metallic glasses [23–25].

The bulk modulus calculated as a function of r_s parameter is shown in Fig. 2 for three different values of the correlation energies. We can see that the magnitude of the bulk modulus decreases with the increase in the electron density parameter r_s. We also recognize that, although the bulk modulus calculated by the jelium model follows the general trend, it overestimates the values of bulk modulus obtained from Eq. 1. The agreement between the two calculations increases if the magnitude of the bulk modulus calculated by the jelium model is multiplied by a certain factor. An example is shown in Fig. 2, when such a factor is 0.4 (the value of $\beta = 0.3$ was used). The important point of this comparison is in to recognize the physical essence that is behind the results shown in Fig. 1. The proportionality and the trend shown in Fig. 2 indicate that the bulk modulus of bulk metallic glasses is determined essentially by the electron density analogously to the case of elemental metals [13, 26, 27]. As shown in Fig. 3, for the case of dB/dP, the values calculated from Eq. 1 is dispersed when plotted as a function of r_s. However, we recognize that the majority of the data are distributed around $dB/dP \approx 5$. It is interesting to note that for elemental metals, the experimental values of dB/dP are distributed in the range $dB/dP \approx 5-6$, irrespective the values of r_s [15]. From Fig. 3 we can also see that, in the range of r_s of interest, the calculated value of dB/dP by the jellium model is almost constant. These observations

reinforce once again the importance of the electron density in the mechanical properties of bulk metallic glasses.

Before the development of bulk metallic glasses, studying mechanical properties of amorphous metals was difficult, because most of the samples were obtained in the form of thin films. Reflecting this short history of research on bulk metallic glasses, there are only few measurements on the pressure dependence on the elastic properties. The data available [12] are shown in Fig. 1. There it has been also shown that the model given by Eq. 1 reproduces quite well the available experimental data. Further experimental studies are required to check the validity of Eq. 1. Such experimental studies will provide insights to understand better the physical background that is behind the result shown in Fig. 2.

Conclusions

Bulk modulus is an important quantity that characterizes the mechanical properties of the materials. In the present article, it is shown that the bulk modulus and its pressure derivative of the bulk metallic glasses can be estimated from the values of the constituent elements and their compositions. It has been shown that the predicted values from the model are in good agreement with the experimental data. Some predictions waiting for experimental verifications have been also given. The physical background of the method has been discussed based on the jellium model of metals. It is suggested that the mechanical properties of bulk metallic glasses is determined essentially by the electron density analogously to the case of elemental metals.

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