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Citation: *J. Appl. Phys.* **109**, 053520 (2011); doi: 10.1063/1.3549819

View online: <http://dx.doi.org/10.1063/1.3549819>

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# Characteristic free volumes of bulk metallic glasses: Measurement and their correlation with glass-forming ability

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(Received 20 November 2010; accepted 16 December 2010; published online 15 March 2011)

A convenient method is proposed for the measurement of the characteristic free volumes, viz., the amount of excess free volume annihilation in structural relaxation  $V_{f-sr}$  and the amount of new free volume production in glass transition  $V_{f-gt}$  of bulk metallic glasses (BMGs) by thermal dilation (DIL) test. Through the DIL tests, the characteristic free volumes are found to be sensitive to the change of glass forming ability (GFA). The  $Pd_{40}Cu_{30}Ni_{10}P_{20}$  BMG has a quite small  $V_{f-sr}$ . For a series of Fe–Cr–Mo–C–B–(Er) BMGs,  $Fe_{48}Cr_{15}Mo_{14}C_{15}B_6Er_2$  with the largest GFA is identified to have the largest  $V_{f-gt}$  and smallest  $V_{f-sr}$ . The correlation between  $V_{f-sr}$  and the squares of critical diameters of these iron-based BMGs can be fitted as a negative exponential function with high accuracy. © 2011 American Institute of Physics. [doi:10.1063/1.3549819]

## I. INTRODUCTION

Glass forming ability (GFA) is very important to understand the formation of bulk metallic glasses (BMGs) and the development of new BMGs.<sup>1–5</sup> A glass forming liquid or amorphous solid with higher density compared to their crystalline counterpart is found to have higher GFA.<sup>6,7</sup> This phenomenon can be explained by Cohen and Turnbull's free volume model (FVM).<sup>8–12</sup> The quantitative relationship between the free volume and GFA, however, has not yet been revealed due to the complexity and inaccuracy of the traditional free volume measurement methods.

No matter how complex the original definition of free volume by Cohen and Turnbull is,<sup>8,9</sup> the free volume of BMGs is actually a part of the volume and changes with temperature, i.e., the free volume being released out in structural relaxation<sup>10,11</sup> and reproduced in glass transition.<sup>11,12</sup> In addition, the changing volume of the isotropic BMGs in the heating process is threefold of the dimension change of BMGs themselves. It is thus possible to measure the free volume change by dilation (DIL) test<sup>13–17</sup> from which the thermal expansion of BMGs can be identified. In tandem with this, a method to quantitatively and conveniently measure the two types of characteristic free volumes, viz., the amount of excess free volume annihilation in structural relaxation  $V_{f-sr}$  and the amount of new free volume production in glass transition  $V_{f-gt}$  is proposed based on the DIL test. Further, the correlations between the characteristic free volumes and GFA for a series of Fe–Cr–Mo–C–B–(Er) BMGs are investigated.

## II. EXPERIMENT

A series of Fe–Cr–Mo–C–B–(Er) (Ref. 18) and  $Pd_{40}Cu_{30}Ni_{10}P_{20}$  (Ref. 19) rods were prepared by copper-mold casting. The amorphous states of the samples were confirmed by x-ray diffraction (Bruker D8). The glass transition temperature  $T_g$ , crystallization temperature  $T_x$  and liquid temperature  $T_l$  were measured with differential scanning calorimetry (DSC) (Setaram SETSYS Evolution 1750). The rods of Fe and Pd BMGs with the lengths of 25 mm were tested with DIL (Netzsch DIL 402C) in argon atmosphere. In the DIL test, the Fe and Pd BMGs were heated up from room temperature to 1073 and 693 K, respectively. The heating rate is 0.0833 K/s. The load applied to the samples is 0.3 N and the instrument resolution is 1.25 nm.

## III. RESULTS AND DISCUSSION

### A. The measurement principle of the characteristic free volumes

Figures 1(a) and 1(b) show the DSC results and the variation of the thermal expansion coefficient  $\alpha$  of a Fe BMG. When the sample is heated up with a constant heating rate, it goes through the different regions in sequence in Fig. 1, which include low temperature relaxation region (Region I),<sup>16,20</sup> high temperature relaxation and glass transition region (II), super-cooled liquid region (III), and crystallization region (IV). Figure 1(a) shows the similarity of the DSC curve and the variation of  $\alpha$  in Region IV ( $T \geq T_{x-on}$ , where  $T_{x-on}$  is the onset crystallization temperature measured by DSC). In the crystallization process, the atomic rearrangement from disorder to order results in the simultaneous energy release and volume shrinkage. The exothermic peak is one-to-one corresponding to the valley of  $\alpha$ , which reflects the volume shrinkage sensitively.

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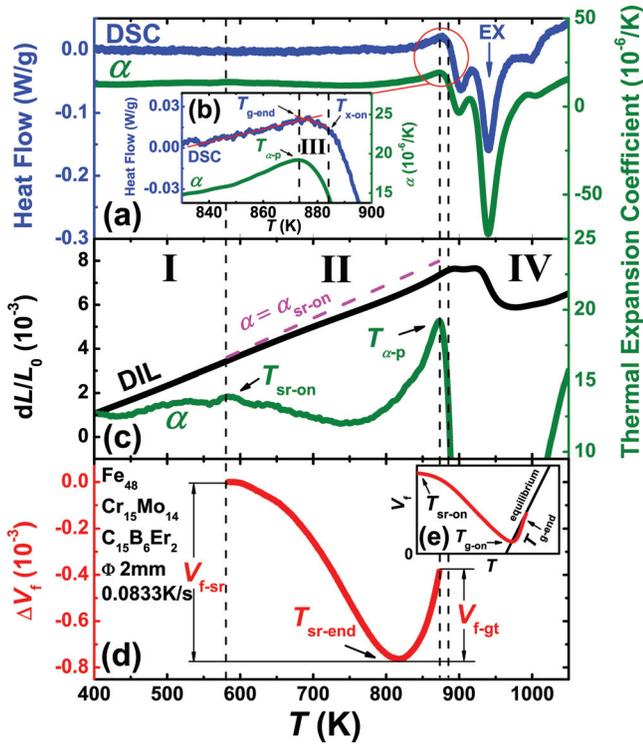


FIG. 1. (Color online) (a) DSC and the thermal expansion coefficient  $\alpha$  curves of Fe BMG; (b) enlarged region around glass transition area; (c) DIL and  $\alpha$  variation (the dashed line is the hypothesis DIL curve); (d) the corresponding  $\Delta V_f(T)$  vs  $T$  curve; (e) free volume change with temperature and the equilibrium line generated based on Eqs. (2) and (3).

As shown in Fig. 1(b),  $T_{g-end}$  in the DSC curve measured by the tangent method is 875.5 K.  $T_{x-on}$  is 884.4 K. The peak temperature of  $\alpha$  curve  $T_{\alpha-p}$  is 872.9 K.  $T_{g-end}$  is very close to  $T_{\alpha-p}$ . According to the FVM, the glass transition is a process where the free volume of glass solid goes out of the free volume of liquid equilibrium and continuously struggles for the attainment of the equilibrium in the continuous heating process.<sup>11,12</sup> This sudden free volume production is not obvious in the DIL curve as shown in Fig. 1(c), as it is overlapped with the comparatively large linear thermal expansion.<sup>13–16</sup> Although it is clearly reflected by the rapid growth of the  $\alpha$  curve, viz., the derivative of the DIL curve. Once the glass transition is completed, the sample enters into the super-cooled liquid region (Region III,  $T_{g-end} \leq T \leq T_{x-on}$ ), and the value of  $\alpha$  decreases quickly as the viscosity reduces dramatically with temperature in this region. Consequently, a peak of  $\alpha$  occurs and the peak temperature corresponds to the temperature at which it is the ending of glass transition and the beginning of the super-cooled liquid region.

Figure 1(c) shows another maximum point at  $T_{sr-on}$  in the  $\alpha$  curve. Before  $T_{sr-on}$ , it is Region I in which the free volume has a minimal change.<sup>16,20</sup> After  $T_{sr-on}$ , it is the high temperature relaxation<sup>10,11</sup> and glass transition region<sup>11,12</sup> (Region II). The high temperature relaxation is the main structural relaxation involving the annihilation of the quench in excess free volume. If there is no high temperature relaxation and glass transition, the sample will expand linearly at a constant slope  $\alpha_{sr-on}$ , in the way of the hypothesis DIL curve

as shown in Fig. 1(c). Hence, the change of free volume with temperature can be calculated by integrating the difference between  $\alpha(T)$  and  $\alpha_{sr-on}$  or simply by subtracting the hypothesis DIL curve from the experimental one in the following:

$$\begin{aligned} \Delta V_f(T) &= 3 \int_{T_{sr-on}}^T [\alpha(T) - \alpha_{sr-on}] dT \quad (T_{sr-on} \leq T \leq T_{\alpha-p}) \\ &= 3 \left( \frac{dL(T)}{L_0} \Big|_{\text{experiment}} - \frac{dL(T)}{L_0} \Big|_{\text{hypothesis}} \right). \end{aligned} \quad (1)$$

By doing so, the free volume annihilation and production can be determined and clearly shown in Fig. 1(d), just like what is shown in Fig. 1(e), which is the sketch of free volume change as a function of temperature according to the following equations:

$$\frac{dV_f}{dT} = -\frac{C V_f^2}{s \gamma v^*} \exp \frac{-E_f}{KT} \left( \exp \frac{-\gamma v^*}{V_f} - \exp \frac{-B}{T - T_0} \right), \quad (2)$$

$$\frac{V_{fe}}{\gamma v^*} = \frac{T - T_0}{B}, \quad (3)$$

$$\Delta C_p = \frac{\beta}{\gamma v^*} \frac{dV_f}{dT}. \quad (4)$$

Based on FVM,<sup>9–12</sup> Eq. (2) is the kinetics differential equation of the free volume annihilation and production. In Eq. (2),  $V_f$  is the free volume per atomic volume,  $s$  is the heating rate,  $E_f$  is the relaxation activation energy,  $T_0$  is the Vogel-Fulcher temperature, and  $C$ ,  $B$ , and  $\gamma v^*$  are constants. Equation (3) is a linear equation of the free volume in the thermodynamic equilibrium ( $V_{fe}$ ) in a narrow region around  $T_g$ .

In addition, Fig. 1(e) is an illustration of the relationship of  $V_f$  and  $T$ . The specific free volume value can be determined via the numerical fitting of Eqs. (2) and (4).<sup>10–12</sup> Equation (4) is based on the assumption that the energy change is caused by the change of free volume. In Eq. (4),  $\Delta C_p$  and  $\beta$  are the specific heat and the proportional coefficient, respectively. To find the final specific free volume value (referring to the zero free volume), there needs to be eight parameters ( $\Delta C_p$ ,  $\beta$ ,  $C$ ,  $s$ ,  $E_f$ ,  $B$ ,  $\gamma v^*$ ,  $T_0$ ). Obviously, this method is a bit complicated. Further, it is difficult to get a reference state of the absolute zero free volume to verify the fitting results.<sup>4,11</sup> On the contrary, Fig. 1(d) has a definitive reference state, which is the free volume contained in the sample before high temperature relaxation. The value of  $\Delta V_f(T)$  shown in Fig. 1(d) is negative, which means the free volume above  $T_{sr-on}$  is smaller than that at  $T_{sr-on}$ . No matter that it refers to the state of the absolute zero free volume [Fig. 1(e)] or the state of the unreleased free volume [Fig. 1(d)], the regularity of the free volume change with temperature will not change and only the vertical coordinate translates. Therefore, the two significant characteristic values of the free volume change,  $V_{f-sr}$  and  $V_{f-gt}$ , can be defined to quantify the amount of excess free volume annihilation in structural relaxation and the amount of new free volume production during glass transition process. Their quantifications are shown in Fig. 1(d). In addition,  $V_{f-sr}$  and  $V_{f-gt}$  are

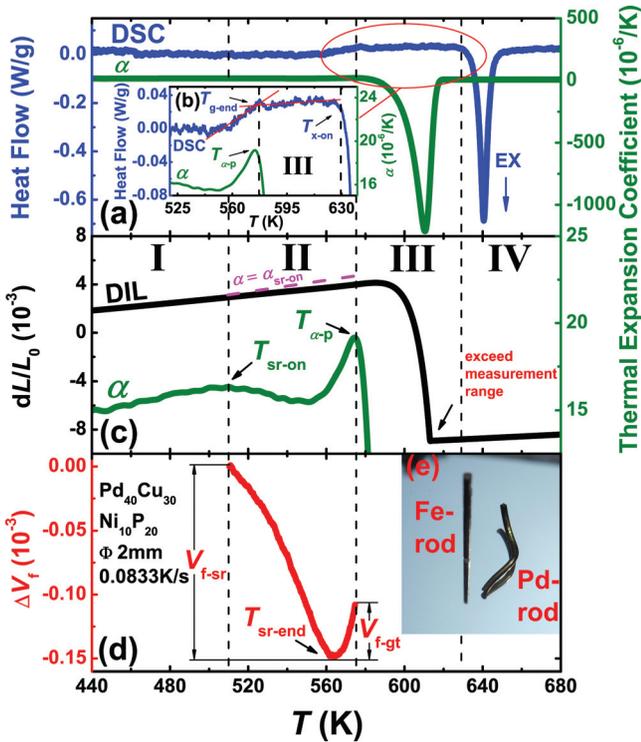


FIG. 2. (Color online) (a) DSC and the thermal expansion coefficient  $\alpha$  curve of Pd BMG; (b) enlarged region around glass transition area; (c) DIL and  $\alpha$  curve. (the dashed line is the hypothesis DIL curve); (d) the corresponding  $\Delta V_f(T)$  vs  $T$  curve; (e) Fe and Pd rods after DIL tests.

calculated as the intercepts from the left ( $T = T_{sr-on}$ ) and the right ( $T = T_{\alpha-p}$ ) highest points to the lowest point ( $T = T_{sr-end}$ ), respectively. The  $T_{sr-end}$  corresponds to the temperature at the end of the structural relaxation as well as the onset of glass transition  $T_{g-on}$ , as shown in Fig. 1(e). However, the two significant types of characteristic free volumes can be obtained accurately and conveniently despite of the fact the specific free volume value is unknown.

Figure 2 is the process to obtain the characteristic free volume of Pd BMG. Compared to the small viscous flow of

Fe BMG (the length shrinkage is about 60  $\mu m$ ), the viscous flow of Pd BMG is quite violent (length shrinkage  $> 2$  mm) due to its low strength and the large Region III.<sup>19</sup> Figure 2(c) shows the sample length shrinkage exceeding the instrument measurement range (250  $\mu m$ ) at above 613 K. The completion of crystallization cannot be observed in the  $\alpha$  curve. Hence, unlike what is shown in Fig. 1(a), the similarity of DSC and the  $\alpha$  curves in Region IV cannot be observed in Fig. 2(a). Figure 2(e) shows that the Fe rod after measurement can still stand vertically, whereas the Pd rod bends significantly. However, the violent viscous flow in Region III does not affect the measurement of  $\Delta V_f(T)$  in Region II. The regularity of Fig. 2(d) is still consistent with the FVM.

The  $V_{f-sr}$  of Pd BMG ( $0.150 \times 10^{-3}$ ) is much smaller than that of Fe BMG ( $0.765 \times 10^{-3}$ ). The GFA of Pd<sub>40</sub>Cu<sub>30</sub>Ni<sub>10</sub>P<sub>20</sub> (with the critical diameter  $D_c$  of 72 nm),<sup>19</sup> however, is far larger than that of Fe<sub>48</sub>Cr<sub>15</sub>Mo<sub>14</sub>C<sub>15</sub>B<sub>6</sub>Er<sub>2</sub> ( $D_c = 12$  nm).<sup>18</sup>  $V_{f-sr}$  is thus related to GFA.

**B. Correlation between the characteristic free volumes and GFA**

To investigate the correlation between the characteristic free volume and GFA, a series of Fe–Cr–Mo–C–B–(Er) BMGs<sup>18</sup> with a similar composition but different  $D_c$  were used in the tests. As shown in Figs. 3(a1)–3(i1) and 3(a2)–3(i2), the  $\alpha$  curve and  $T_{\alpha-p}$  have a good agreement with DSC and  $T_{g-end}$ , respectively. However, it is difficult to observe how the free volume changes based on the DIL and  $\alpha$  curves shown in Figs. 3(a3)–3(i3), although a clear variation trend can be observed in Figs. 3(a4)–3(i4), which were obtained by the aforementioned method. The free volume production in glass transition could hardly be observed in Figs. 3(a4)–3(d4), but clearly appears in Figs. 3(e4)–3(i4). The compositions with large GFA ( $D_c > 4$  mm) show a clear  $V_{f-gt}$ . The Fe<sub>48</sub>Cr<sub>15</sub>Mo<sub>14</sub>C<sub>15</sub>B<sub>6</sub>Er<sub>2</sub> sample has the largest  $V_{f-gt}$  ( $0.240 \times 10^{-3}$ ) and its GFA is also the largest ( $D_c = 12$  nm), which further shows the clearest glass transition related to the best GFA in those with similar compositions. On the other hand, the larger the

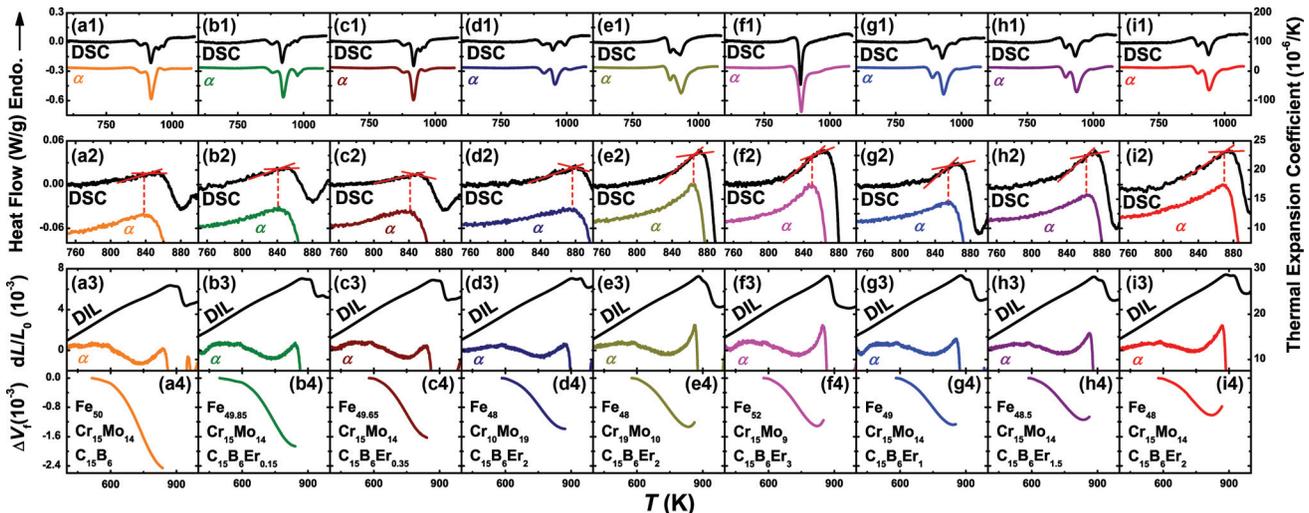


FIG. 3. (Color online) (a1)–(i1) DSC and the  $\alpha$  curves of Fe–Cr–Mo–C–B–(Er) BMGs rods with the diameter of 1.0 mm. The heating rate of DSC and DIL is 0.0833 K/s; (a2)–(i2) enlarged regions around the glass transition areas of (a1)–(i1); (a3)–(i3) DIL and the  $\alpha$  curves; (a4)–(i4) the corresponding  $\Delta V_f(T)$  vs  $T$  curves.

TABLE I. Summary of the thermal analyses by DSC [ $\Delta T_x = T_x - T_g$ ,  $T_{rg} = T_g/T_l$ ,  $\gamma = T_x/(T_g + T_l)$ ,  $\gamma_m = (2T_x - T_g)/T_l$ ],  $V_{f-sr}$  and  $D_c$  of a series of Fe–Cr–Mo–C–B–(Er) BMGs (diameter: 1.0 mm; heating rate: 0.3333 K/s for DSC and 0.0833 K/s for DIL).

Alloy	Composition	$T_g$ (K)	$T_x$ (K)	$T_l$ (K)	$T_x$ (K)	$T_{rg}$	$\gamma$	$\gamma_m$	$V_{f-sr} (\times 10^{-3})$	$D_c$ (mm)
a	Fe <sub>50</sub> Cr <sub>15</sub> Mo <sub>14</sub> C <sub>15</sub> B <sub>6</sub>	841	863	1531	22	0.549	0.364	0.578	2.460	1.5
b	Fe <sub>49.85</sub> Cr <sub>15</sub> Mo <sub>14</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>0.15</sub>	839	864	1528	25	0.549	0.365	0.582	1.862	2
c	Fe <sub>49.65</sub> Cr <sub>15</sub> Mo <sub>14</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>0.35</sub>	830	865	1537	35	0.540	0.365	0.586	1.626	3
d	Fe <sub>48</sub> Cr <sub>10</sub> Mo <sub>19</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>2</sub>	840	895	1557	55	0.540	0.373	0.610	1.394	4
e	Fe <sub>48</sub> Cr <sub>19</sub> Mo <sub>10</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>2</sub>	845	890	1530	45	0.552	0.375	0.611	1.338	5
f	Fe <sub>52</sub> Cr <sub>15</sub> Mo <sub>9</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>3</sub>	842	885	1510	43	0.558	0.376	0.615	1.316	6
g	Fe <sub>49</sub> Cr <sub>15</sub> Mo <sub>14</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>1</sub>	823	884	1513	61	0.544	0.378	0.625	1.280	7
h	Fe <sub>48.5</sub> Cr <sub>15</sub> Mo <sub>14</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>1.5</sub>	830	889	1518	59	0.547	0.379	0.625	1.140	9
i	Fe <sub>48</sub> Cr <sub>15</sub> Mo <sub>14</sub> C <sub>15</sub> B <sub>6</sub> Er <sub>2</sub>	843	895	1515	52	0.556	0.380	0.625	1.017	12

$D_c$ , the smaller the free volume annihilation is. Compared to the relation between  $V_{f-gt}$  and GFA, this observation is more accurate. From Table I, it is found that only  $\gamma$ , (Ref. 1)  $\gamma_m$  and  $V_{f-sr}$  (Ref. 2) have a good correlation with  $D_c$ . However,  $\gamma$  and  $\gamma_m$  change slightly when  $D_c$  changes greatly. The relative changes of  $\gamma$  and  $\gamma_m$  corresponding to the largest and smallest  $D_c$  are  $(0.380 - 0.364)/0.364 = 4\%$  and  $(0.625 - 0.578)/0.578 = 8\%$ , respectively. On the other hand, the relative change of  $V_{f-sr}$  is  $(1.017 \times 10^{-3} - 2.460 \times 10^{-3})/(2.460 \times 10^{-3}) = -59\%$ . Therefore,  $V_{f-sr}$  is much more sensitive to the change of GFA for those Fe-based BMGs.

From the perspective of thermal conduction, the volume of rod with the critical diameter (proportional to  $D_c^2$ ) represents the GFA more appropriately than the lateral area (proportional to  $D_c$ ). Figure 4 shows the correlation between  $D_c^2$  and  $V_{f-sr}$ . The data are fitted by a negative exponential function with the regression coefficient  $R^2$  up to 0.99. In addition, the correlation between  $D_c$  and  $V_{f-sr}$  can also be fitted with a high accurate negative exponential function ( $R^2 = 0.98$ ). Actually, if the approximate inverse proportion relation between the critical cooling rate  $R_c$  and  $D_c^2$  [ $R_c = 4 \times 10^3 / D_c^2$  (mm)], derived by Lin and Johnson,<sup>5</sup> is used, the fitting function is quite similar to the results obtained by Park and Kim.<sup>21</sup> In their fitting function, the independent variable is  $(V_l - V_g)/V_l$ , where  $V_l$  and  $V_g$  is the specific volume at the

melting temperature and the glass transition temperature, respectively.

The measurement process is actually an isochronal annealing. Under the condition of the same heating rate, the degree of the excess free volume annihilation in each sample is the same. Therefore, the smaller  $V_{f-sr}$  means the smaller excess free volume contained in the sample. According to Table I, the  $T_g$  and  $T_l$  of those samples with the diameter of 1.0 mm have a slight difference.  $(T_l - T_g)$  is  $(690 \pm 20)$  K. In this case, the cooling rate for preparing these samples can be seen as the same.<sup>5</sup> Under the condition of the same cooling rate, the smaller excess free volume of the amorphous solid state means the smaller free volume of liquid metal.<sup>10</sup> On the one hand, according to FVM, the smaller free volume of liquid results in higher liquid viscosity<sup>8,9</sup> and therefore lower atomic mobility, as well as the more stabilized liquid phase.<sup>1-7,21-23</sup> On the other hand, the glass forming liquid with a smaller free volume has a smaller entropy of fusion and consequently smaller gradient in  $\Delta G$  (Gibbs free energy difference between the super-cooled liquid and the crystal) at the melting point, which results in a lower driving force for crystallization and therefore a greater GFA.<sup>3,4,22</sup> In short, the viscosity and  $\Delta G$ , the two dominant factors of GFA, are related to each other via the free volume of liquid. After fast cooling, the free volume of liquid is frozen into the excess free volume of the amorphous solid state, and finally released out as  $V_{f-sr}$ , which can be measured by DIL test.

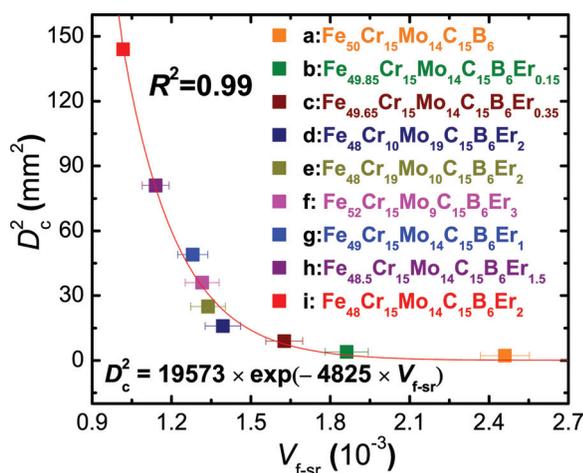


FIG. 4. (Color online) Correlation between the GFA (represented by the square of the critical diameter) and the  $V_{f-sr}$ .

#### IV. CONCLUSIONS

A convenient method for the measurement of the characteristic free volumes of BMGs is proposed in this paper. Through measurement and testing, it is found that: (a) the free volume annihilation in structural relaxation  $V_{f-sr}$  is very sensitive to GFA; (b) Pd<sub>40</sub>Cu<sub>30</sub>Ni<sub>10</sub>P<sub>20</sub> BMG has a quite small  $V_{f-sr}$ ; (c) The correlation between  $V_{f-sr}$  and  $D_c^2$  for a series of Fe–Cr–Mo–C–B–(Er) BMGs can be fitted by a negative exponential function with high accuracy. The method developed can be used to study the relationship between structure and properties of BMGs for its simplification, quantification and accuracy.

#### ACKNOWLEDGMENTS

This research was conducted with financial support from the Science and Technology Foundation of Shenzhen China

(CXB200903090012A) and the Two Hundred Plans for Talent Station of Shenzhen (Shenfu [2008] No. 182).

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