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Effect of randomization on ductility of brittle Fe–Si–B glass

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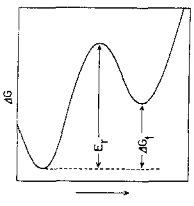
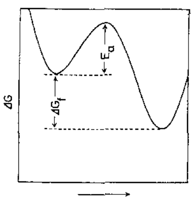
Using the Vickers indentation method, a critical plastic deformation energy, E_c, of collapse was proposed to be a convenient measurement to determine the ductility. It was found that the randomization increases the E_c value of a brittle Fe–Si–B alloy glass which is cooled slowly. Namely, the randomization enhances the ductility of the metallic glass.

1. Introduction
Liquid-quenched Fe–Si–B alloy glasses are practical soft magnetic materials [1–3]. Although rapidly cooled metallic glasses are, in general, ductile, slowly cooled or aged glasses are brittle. From the engineering point of view, it is important to reduce the brittleness homogeneously. Randomization is effective for homogeneous disordering [4] and softening [5] of the rapidly cooled glasses [6, 7]. Thus, randomization may improve the ductility of the brittle metallic glass, whereas ageing, which brings the structure close to equilibrium (see Table I), reduces ductility.

The stress intensity factor, K_{Ic}, has been used as a standard to evaluate the brittleness [8–10]. However, it is difficult to prepare test specimens to measure this parameter from brittle materials by a conventional method. Thus, a new method has been suggested by means of a standard hardness tester for different materials [11–13]. When collapses form around a Vickers indentation under a certain load, it is suggested that a critical load, a critical diagonal and a critical deformation energy of collapse, can be defined. They are relatively easy to measure on small samples such as metallic glasses. The purpose of the present work was to evaluate the effect of the randomization on the ductility (brittleness) for the Fe–Si–B alloy glasses in terms of the critical deformation energy.

TABLE I Schematic concept of randomizing and ageing

	Randomizing	Ageing
Structure change	Homogenization	
	Destabilization Disordering Random distribution	Stabilization Ordering Relaxation
Energy	Collision energy	Thermal energy
Rate process		

2. Experimental procedure
The master alloy of Fe–10 at % Si–15 at % B was prepared from high-purity powders of pure iron (99.9%), silicon (99.99%) and boron (99.5%). The powders were mixed, pelletized, sintered and melted under an argon atmosphere. Liquid-quenched glassy foils were prepared using a twin-type piston–anvil apparatus [14–16]. This apparatus was constructed to remelt, by an infrared furnace at about 1700 K, and then to quench rapidly, the molten sample under an Ar–5 vol % H_2 atmosphere.

The randomization was performed by a peening apparatus operated under the following conditions [3–5]. The nozzle diameter was 8.0 mm, the velocity of air at the nozzle was 195.5 m s^{-1}, the distance between the nozzle and the specimen was 20 mm, and the peening angle was 30° to the specimen surface. The steel balls were made of SUJ 2 steel [$0.95 < C(\text{mass }\%) < 1.10$, $0.15 < Si(\text{mass }\%) < 0.35$, $1.30 < Cr(\text{mass }\%) < 1.60$, HRC (Rockwell hardness C scale) = 64]; their mean diameter and weight were 0.4 ± 0.1 mm and 0.68 mg, respectively, and they were supplied at a rate of 18.1 mg mm^{-2} s^{-1}.

The structure of the sample was examined by means of X-ray diffraction. The crystalline diffraction peaks were not observed for the glassy samples. A precise evaluation of ductility was performed with a standard Vickers microhardness tester.

3. Results and discussion
3.1. Evaluation of brittleness
Fig. 1a and b show an optical photograph and schematic profile respectively, of a Vickers indentation

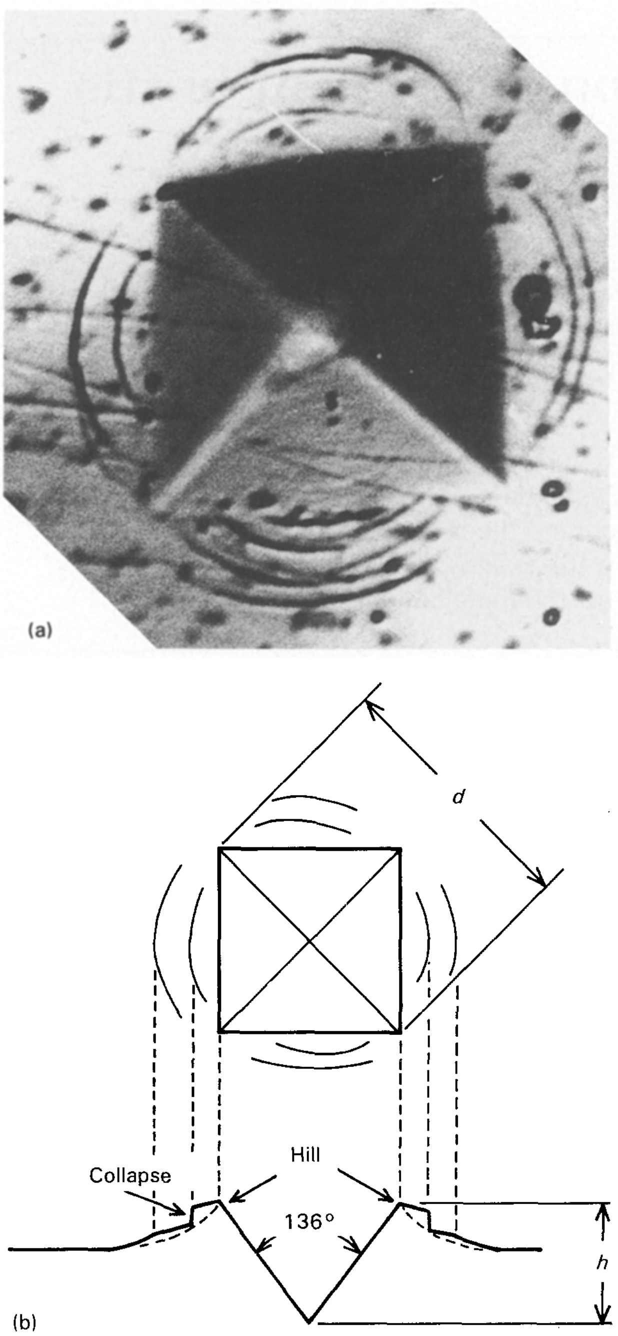


Figure 1 Optical photograph and schematic profile of a Vickers indentation with an accompanying collapse of the brittle Fe–Si–B glass which is cooled slowly.

with an accompanying collapse with the brittle Fe–Si–B glass which was cooled slowly. The collapse existed at the edge of the hill. The shape of the collapse is apparently different from those of other samples previously reported [11–13]. When a collapse is found outside the indentation, the critical values of d and P are defined. The critical diagonal, d_c^{mid}, and critical load, P_c^{mid}, were defined as the midpoint between the minimum and maximum values [13], i.e.

$$d_c^{mid} = (d_c^{min} + d_c^{max})/2 \quad (1)$$

$$P_c^{mid} = (P_c^{min} + P_c^{max})/2 \quad (2)$$

When one of five indentations exhibits a collapse at a certain load, the minimum load, P_c^{min}, and minimum diagonal, d_c^{min}, are defined. P_c^{max} and d_c^{max} are the maximum load and maximum diagonal, respectively, for which one of five indentations is collapse-free. Therefore, a critical deformation energy, E_c^{mid}, of collapse is suggested here as a new parameter to evaluate brittleness. The deformation energy can be expressed by the following equation [13]

$$E_c^{mid} = 0.0477\ P_c^{mid} d_c^{mid} \quad (3)$$

3.2. Randomization

The critical values, d_c^{mid}, P_c^{mid} and E_c^{mid}, of the randomized Fe–10 at % Si–15 at % B alloy glasses are summarized in Table II. Fig. 2 shows changes in d_c^{mid}, P_c^{mid} and E_c^{mid} against the randomization time, t_r. The critical values for the randomized glasses are larger than that for the as-quenched glass which is cooled slowly and is brittle. The longer the randomization time, the larger the critical values become; namely, randomization enhances ductility.

If the energy involved in randomization is taken to be the collision energy, a rate process may be applied. Based on the rate process [3–5], the volume-fraction change, X, is generally expressed by the following equation in relation to the randomization time, t_r

$$X = 1 - \exp(-kt_r^n) \quad (4)$$

here, k and n are constants. X is assumed to express

$$X = [(E_c^{mid} - {}^0E_c^{mid})/({}^mE_c^{mid} - {}^0E_c^{mid})] \quad (5)$$

TABLE II The critical values of the randomized Fe–10 at % Si–15 at % B alloy glasses

Randomizing time (s)	P_c^{mid} (gf)	d_c^{mid} (μm)	E_c^{mid} (erg)
50	100	11.545	5.397
50	100	11.715	5.476
100	100	11.680	5.460
100	125	13.270	7.754
200	125	12.725	7.435
200	125	13.078	7.641
500	125	13.103	7.656
500	175	14.988	12.260
1000	175	14.623	11.962
1000	150	13.358	9.366

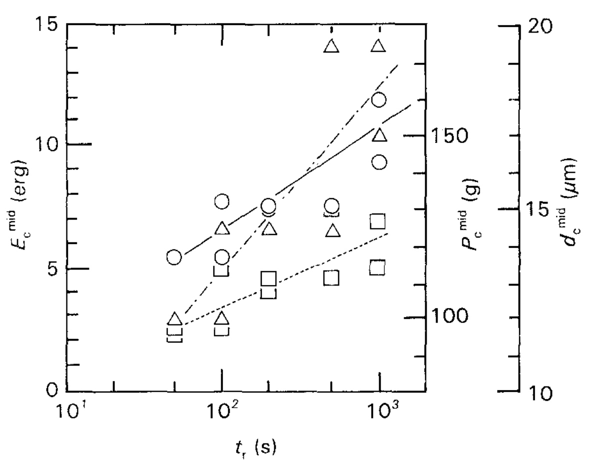


Figure 2 Change in (□) d_c^{mid}, (△) P_c^{mid}, and (○) E_c^{mid} with the randomizing time, t_r.

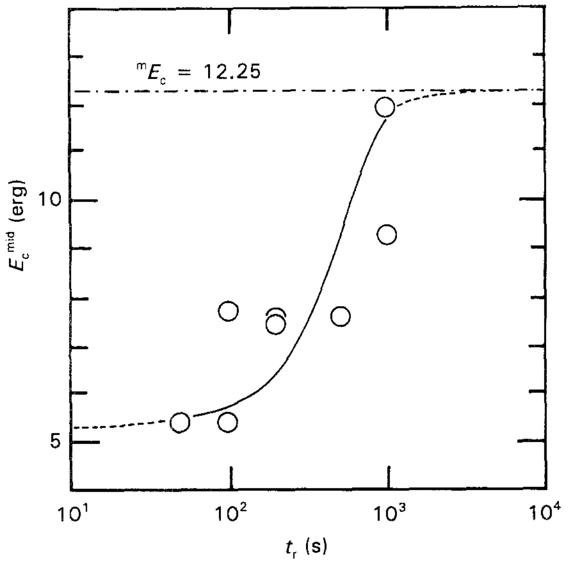


Figure 3 Change in E_c^{mid} with the randomizing time, t_r, of liquid-quenched Fe–10 at % Si–15 at % B alloy glasses. $^{m}E_c^{mid}$ is the E_c at extremely long randomizing time.

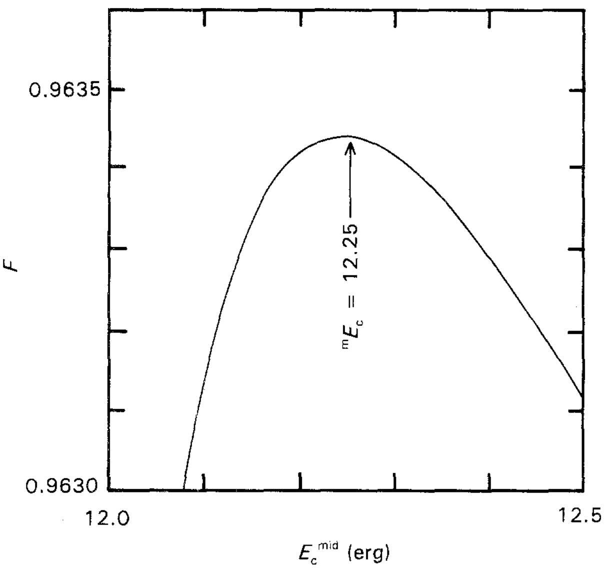


Figure 4 Change in correlation coefficient, F, with E_c^{mid} for Fe–10 at % Si–15 at % B alloy glass. $^{m}E_c^{mid}$, which is the E_c at extremely long randomizing time, is obtained at the maximum value of F.

where $^{m}E_c^{mid}$ and $^{0}E_c^{mid}$ are the E_c^{mid} of glass randomized for extremely long periods of time and of glass before randomization, respectively. E_c^{mid} of the randomized glass approaches the estimated $^{m}E_c^{mid}$ value in Fig. 3. When the correlation coefficient, F, of Equation (5) is maximum, as shown in Fig. 4 [17], the estimated $^{m}E_c^{mid}$ value is 12.25 erg. From these results, X is expressed by the following equation for Fe–Si–B alloy glasses (see Fig. 5)

$$\log_{10}[-\ln(1-X)] = n\log_{10}t_r + \log_{10}k \quad (6)$$

Equation (6) is plotted as the solid line in Fig. 5. This linear plot of Equation (8) confirms the assumption of the rate process (see Equation (4)). Therefore E_c^{mid} can

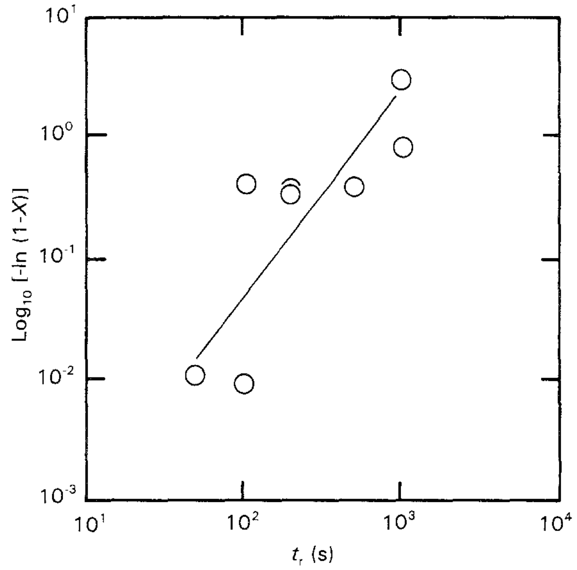


Figure 5 Linear plot of $\log_{10}[-\ln(1-X)]$ against t_r, $X = [(E_c^{mid} - {}^{0}E_c^{mid})/({}^{m}E_c^{mid} - {}^{0}E_c^{mid})]$, when $^{m}E_c^{mid}$ and $^{0}E_c^{mid}$ are values after and before time randomizing, respectively.

be obtained by controlling the randomization time, as wished.

4. Conclusion

The ductility (brittleness) for the randomized Fe–10 at % Si–15 at % B glass has been evaluated by means of a microhardness tester. Based on the rate process, the randomization enhances the ductility of the brittle metallic glass by the use of the critical deformation energy of collapse. The deformation energy can be controlled by the randomization time.

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