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Low-temperature fracture toughness of some iron, nickel-based metallic glass ribbons

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The critical stress intensity factor of $Fe_{40}Ni_{40}B_{20}$, $Ni_{80}Si_{10}B_{10}$ and $Ni_{80}Si_5B_{15}$ metallic glass ribbons was measured at temperatures 4.2 to 300 K. The low-temperature embrittlement for Ni–Si–B glasses was not investigated. The low-temperature embrittlement of $Fe_{40}Ni_{40}B_{20}$ glass depends on the structural relaxation and is connected with a change from vein pattern fracture surface morphology through chevron morphology up to cleavage.

1. Introduction

The measurement of the fracture toughness of metallic glasses and the fractographic analysis of fracture surfaces [1–3] yields information about the local processes of plastic deformation during the failure of these materials. It was shown [4, 5], that the unusual dimensions of metallic glass ribbons pose some problems in the interpretation of the measured values of the fracture toughness. This is caused by the failure under plane stress conditions, when the measured critical stress intensity factor, K_c, does not represent the fracture toughness K_{Ic}, which is defined for failure under plane strain conditions.

It follows from recent papers [6, 7], that some amorphous metallic structures do not become brittle even at very low temperatures. The aim of this work was to observe in more detail, by means of fracture toughness measurements and fractographic analysis, the processes of failure in two types of transition metal-based glasses, which exhibit different behaviour during failure at low temperatures.

2. Experimental detail

The measurements were made on $Fe_{40}Ni_{40}B_{20}$, $Ni_{80}Si_{10}B_{10}$ and $Ni_{80}Si_5B_{15}$ amorphous ribbons with cross-sections of $9.55 \times 0.036\,mm^2$, $10.2 \times 0.023\,mm^2$ and $10.2 \times 0.026\,mm^2$, respectively. The amorphousness of these materials was confirmed during the experiments by a series of X-ray diffraction tests. The strength and K_c measurements, using the tensile test on specimens with a centrally located sharp crack, were made at 4.2, 77, 200 and 300 K; the deformation rate was $5 \times 10^{-5}\,sec^{-1}$. The dimensions of the plastic zone on the crack tip after failure were determined using an optical microscope Neophot 21 and the fractographic analysis was performed on a scanning electron microscope TESLA BS 300. The more detailed characteristics of the samples, the conditions for the formation of a sharp crack by the high-cycled ultrasound loading, and the method of K_c determination from the tensile test record of the specimen with centrally located crack, have been described previously [8].

3. Results

3.1. Fracture toughness and tensile tests

The measured values of the critical stress intensity factor for the $Fe_{40}Ni_{40}B_{20}$ metallic glasses and for both Ni–Si–B alloys in the temperature range 4.2 to 300 K are shown, together with the tensile strength data, in Fig. 1. The bars indicating 95% reliability of the K_c mean values assuming normal distribution, are also shown. Previous measurements [5] have shown that K_c in metallic glasses may depend on the initial crack length. However, in all our measurements the crack was longer than one-tenth and shorter than one-quarter of the sample width. Statistical evaluation showed that in this range of crack lengths no dependence of K_c on crack length is manifested.

The strength of the $Fe_{40}Ni_{40}B_{20}$ and $Ni_{80}Si_5B_{15}$ alloys, as well as the critical stress intensity factor K_c of both Ni–Si–B alloys, show no significant change with decreasing temperature. The hypothesis that the mean values of K_c are the same at various temperatures for both Ni–Si–B alloys was not rejected by a statistical t-test. The measured mean values of K_c for $Ni_{80}Si_{10}B_{10}$ and $Ni_{80}Si_5B_{15}$ are 40 and 45 $MPa\,m^{1/2}$, respectively.

On the other hand, the fracture toughness of $Fe_{40}Ni_{40}B_{20}$ alloy apparently decreases with decreasing temperature from about 63 $MPa\,m^{1/2}$ at 300 K to about 10 to 15 $MPa\,m^{1/2}$ at 4.2 K. It was found that the

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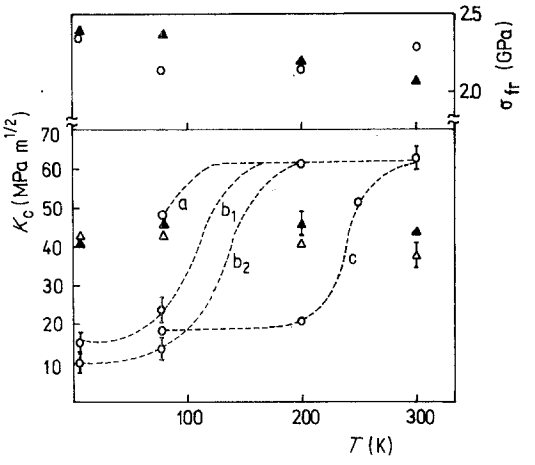


Figure 1 Temperature dependence of the fracture stress and critical stress intensity factor for the (○) $Fe_{40}Ni_{40}B_{20}$, (△) $Ni_{80}Si_{10}B_{10}$ and (▲) $Ni_{80}Si_5B_{15}$ metallic glasses. Curves a, b and c correspond to fracture toughness tests made 5, 14 and 20 months after preparation of $Fe_{40}Ni_{40}B_{20}$ samples.

low-temperature embrittlement of this material depends on the time elapsed from the preparation of material. Fig. 1 shows the dependence of the low-temperature embrittlement assuming that the type of this dependence is similar to that observed in the crystalline metal materials. Curves a, b and c correspond to the measurements made 5, 14 and 20 months after the preparation of the ribbon, respectively. During ageing all samples were kept at room temperature. The initial structural state is the second factor influencing the degree of embrittlement of this material. We noticed that samples measured 14 months after their preparation may be divided (at test temperatures 77 and 4.2 K) into two groups (corresponding to curves b_1 and b_2) with statistically different mean values of K_c. This splitting corresponds exactly to the place from which the sample was taken from the ribbon.

3.2. Fractography

Failure of metallic glasses may be characterized by either ductile (two modes) or cleavage failure mechanism in the plane of maximum shear stresses. At low in descending order of fracture energy, we have:

(a) ductile mechanism in the plane of maximum shear stresses with a typical vein morphology of the fracture surface;

(b) ductile mechanism in the plane of maximum normal stresses with a typical morphology of grooves (chevron pattern);

(c) cleavage in the planes of maximum normal stresses with cleavage morphology.

3.2.1. Ni–Si–B alloys

On both types of these samples and at all temperatures only ductile failure in the plane of maximum shear stresses with smooth fracture surfaces was observed (Fig. 2a, b). Because of very intense shear, the veins may be observed only on the edges.

3.2.2. $Fe_{40}Ni_{40}B_{20}$ alloy

At low temperatures this alloy showed a different failure mode for specimens without an a priori crack (strength test) and for specimens with induced crack (K_c measurement). At strength tests down to 4.2 K the samples failed under the formation of vein morphology, i.e. by the energetically most demanding mechanism in the plane of maximum shear stresses. At low temperatures we observed on these samples only a few regions with chevron morphology near stress concentrators, represented by a cross-sectional decrease (sample necking) or by the presence of a crystallized region (Fig. 3).

On the sample with a sharp crack (Fig. 4), a gradual transition from shear rupture in the plane of maximum shear stresses ($K_c \sim 60\,MPa\,m^{1/2}$, Fig. 4a), through ductile failure in the plane of maximum tensile stresses ($K_c \approx 15$ to $20\,MPa\,m^{1/2}$, Fig. 4b) to cleavage ($K_c = 10\,MPa\,m^{1/2}$ at temperatures of 4.2 K, Fig. 4c) may be observed.

It was also interesting to compare the fracture morphology for different aged samples failed at 77 K. A specimen tested 5 months after its preparation shows a combination of both modes of ductile failure. Both groups of samples tested after 14 months failed through a ductile mechanism in the plane of maximum normal stresses. However, they showed different chevron morphology, as seen from Figs 4b and 5.

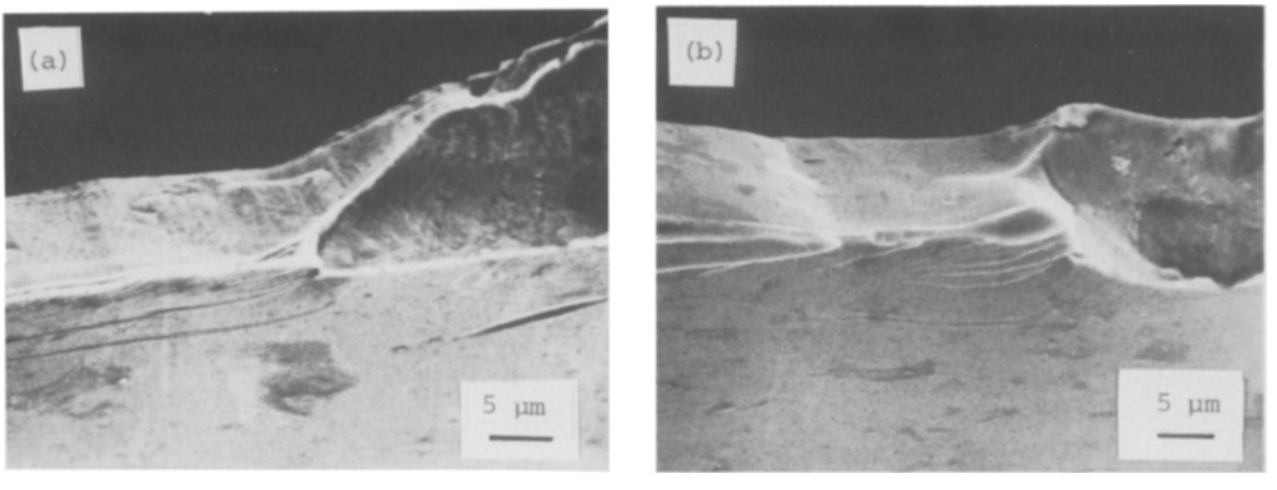


Figure 2 Fracture surface morphology of the $Ni_{80}Si_{10}B_{10}$ metallic glass showing the transition from fatigue to tensile failure. The crack propagated from the right; (a) 300 K, (b) 4.2 K.

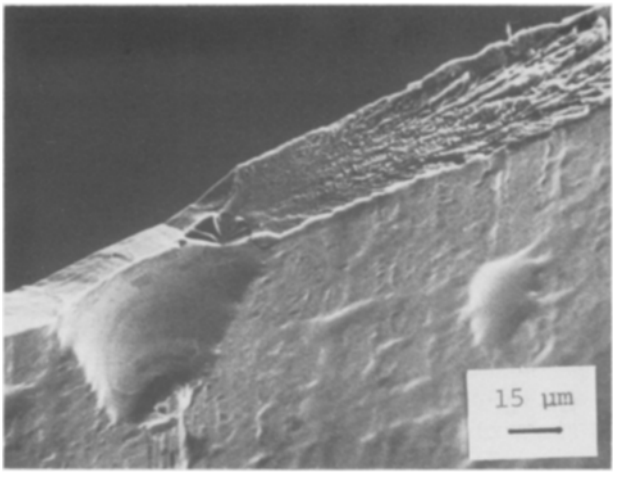


Figure 3 $Fe_{40}Ni_{40}B_{20}$ alloy after failure in a strength test at 77 K. Failure in the plane of maximum normal stresses with chevron morphology could be observed only in a few regions, usually near strong stress concentrators. Failure in the plane of maximum shear stresses with vein morphology predominant, see left side of figure.

3.3. Size of the plastic zone on the crack tip

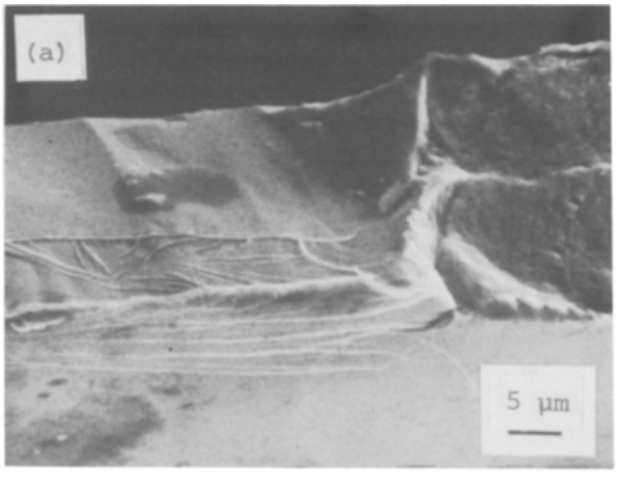
On samples whose failure is preceded by an inhomogeneous shear, the plastic zone on the crack tip is formed through the intense deformation in shear bands, which may be observed under an optical microscope (Fig. 6). During the pile-up of plastic deformation, a system of shear bands is formed on the crack tip; at a greater distance from the crack root these bands may be found near the planes making angle of 45° with the direction of tension loading. Failure results when the crack propagates in one of these shear bands. We measured the width and the length of the plastic zone on the crack tip; the length was defined as the maximum distance reached by bands originating in the crack root. We observed that in the $Ni_{80}Si_{10}B_{10}$ and $Ni_{80}Si_5B_{15}$ samples the width of the plastic zone is independent of the test temperature and is approximately equal to the sample thickness. However, the length of the plastic zone decreases monotonically with decreasing temperature, from 190 μm at 300 K to 40 μm at 4.2 K for $Ni_{80}Si_{10}B_{10}$ and from 105 μm at 300 K to 50 μm at 4.2 K for $Ni_{80}Si_5B_{15}$. Because of the large data scattering (as much as 20%) for various samples at the same test temperature, the results of these measurements should be considered only qualitatively.

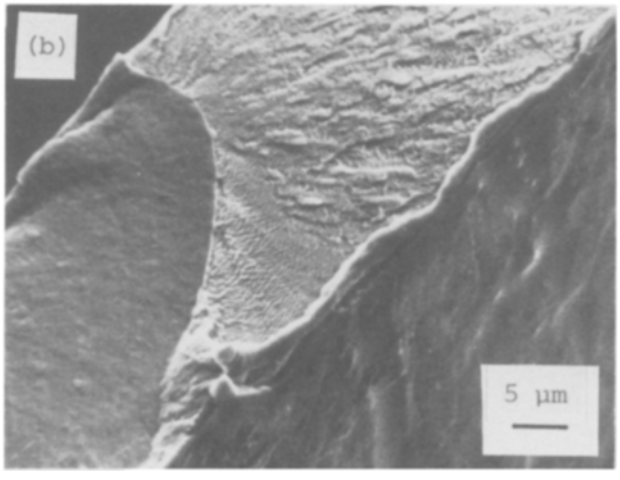
4. Discussion

It may be assumed from [10] that the plastic properties of metallic glasses are (as in crystalline materials) determined by the properties and distribution of some sort of "defects", i.e. such regions in the amorphous structure where the energy required for the displacement of a group of atoms is relatively low. This displacement results in a local shear deformation, which is elastically transferred through the surrounding structure to the surface [10].

A number of definitions of the defects in an amorphous structure has been already proposed [10]. Considering the influence of these defects on the plastic properties, the most interesting conceptions of defects are those based on the fluctuations of both free volume [11] and the invariants of the internal stress tensor on an atomic level [12].

It follows from the proposed micromechanism of plastic deformation in metallic glasses [10], that the more defects an amorphous structure contains, the better is its ability to undergo a plastic deformation. This was confirmed by observations of the amorphous alloy embrittlement after low temperature structural relaxation, e.g. [1], and after hydrogenation [13]. Both





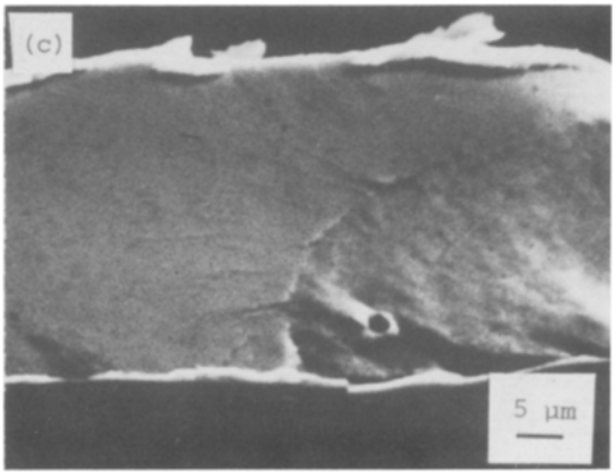


Figure 4 Fracture surface of the $Fe_{40}Ni_{40}B_{20}$ sample after failure in a fracture toughness test showing the transition from fatigue to tensile failure; 14 months after preparation. (a) 200 K, (b) 77 K, sample corresponding to curve b_2 in Fig. 1, (c) at 4.2 K.

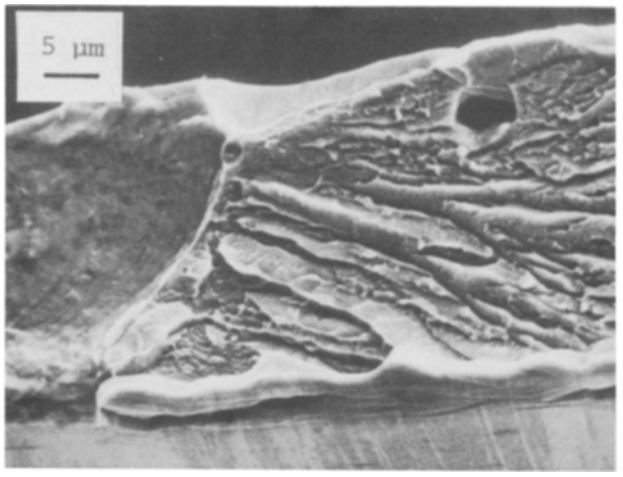


Figure 5 Fracture of the $Fe_{40}Ni_{40}B_{20}$ sample showing the transition from fatigue to tensile failure; 14 months after preparation, at 77 K. Sample corresponds to curve b_1 in Fig. 1.

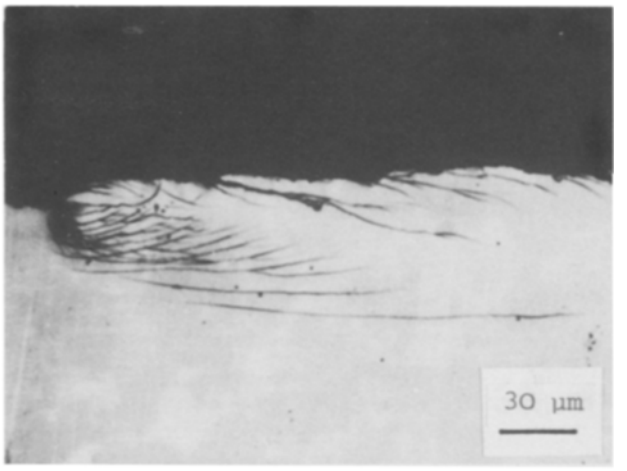


Figure 6 Shear bands forming a plastic zone on the crack tip. $Fe_{40}Ni_{40}B_{20}$ sample after failure at 300 K.

these processes lead to a decrease in the free volume with the resulting decrease in the density of defects [14]. On the other hand, experiments with the bombardment of embrittled structures [15] demonstrate the recovery of plastic properties after irradiation with high neutron doses.

Only few papers have been published on the failure of metallic glasses at low temperatures, with somewhat contradictory results. Bengus *et al.* [6] measured the strength of the $Fe_{40}Ni_{40}B_{20}$ and $Ni_{78}Si_8B_{14}$ alloys down to 4.2 K, but only in an iron-containing alloy was an anomalous decrease of the tensile strength observed, in comparison with the dependence of Young's modulus on temperature. A similar dependence on the same alloy in the temperature range 77 to 300 K was observed by Calvo *et al.* [7]; however, they found no change in the fracture toughness in this temperature range. This result is contradictory to our results shown in Fig. 1. We assume that the explanation may be found in the principal difference between the tensile test and the fracture toughness test. Failure at the tensile test also includes the process of crack nucleation, in which the geometric unevenness (as in Fig. 3) plays a role as a local stress concentrator. Moreover, these narrower regions may be considered to be more brittle, because of the lack of contact with the cooling surface during the casting of the ribbon, caused by a low concentration of the free volume. On the other hand, for the K_c measurements the failure is governed only by the properties of the amorphous structure ahead of the crack tip and is practically uninfluenced by the presence of stress concentrators caused by surface unevenness.

The reasons for the difference in σ_{fr} and K_c dependences for $Fe_{40}Ni_{40}B_{20}$ alloy in this work and that of Calvo *et al.* [7] are, we suppose, the following:

(a) Specimens used by Calvo *et al.* [7] were tested shortly after their preparation in a totally unrelaxed state (even after 30 min annealing at 500 K they showed no tendency to embrittlement) while our specimens were tested as late as 5, 14 and even 20 months after they have been prepared, and therefore some structural relaxation may be reasonably expected. This is also the explanation for the fact, that our K_c-measurement also revealed the low-temperature embrittlement of this alloy;

(b) Our specimens showed probably a much lower surface unevenness, so the crack nucleation at strength tests began at higher stress values, when the whole specimen was in such a stressed state, that the inhomogeneous shear could also take place at low temperatures.

It seems, with respect to the results of the theoretical work [16], that an important role is also played by the deformation rate, $\dot{\varepsilon}$, which might be different in our tests compared to that in [7].

In 1983 the first theoretical work on embrittlement of the amorphous metal structure was published by Steif [16]. This model was based on two assumptions. First, he assumed the validity of the equation for rate of local shear plastic deformation, $\dot{\gamma}$, during the mechanical loading of an amorphous structure, which was derived by Spaepen [17] from atomistic conceptions:

$$\dot{\gamma} = 2\nu \exp\left(\frac{-\Delta G^m}{kT}\right) \exp\left(\frac{-\alpha v^*}{v_F}\right) \sinh\left(\frac{\tau\Omega}{2kT}\right) \quad (1)$$

where ν is Debye frequency, ΔG^m the activation energy of local shear deformation for group of atoms, α a geometrical factor, v^* the critical value of the free volume needed for shear, v_F the mean free volume per one atom, τ the shear stress, k the Boltzmann constant, T the temperature and Ω the volume per atom. The first term in Equation 1 represents the number of elementary deformations in a unit volume per unit time, the second term represents the density of defects and the third term represents the dependence on the applied stress. The second assumption of the Steif model is that the mean free volume per atom, v_f, depends on the hydrostatic stress, σ_{kk}. However, the response of the amorphous structure to the increase of this stress shows a certain time lag, because some time is needed for reordering of atoms. Steif proposed to describe this behaviour by a first-order differential equation:

$$\frac{dv_F}{dt} + \frac{1}{\tau_v}(v_F - v_0) = \tfrac{1}{3}\Omega \frac{\sigma_{kk}}{K\tau_v} \quad (2)$$

where v_0 is the initial free volume without the presence of applied stress, K the bulk modulus and τ_V is the time constant of free volume reordering. Steif solved equations for the uniform tensile loading of a body with stress concentrators and he found that in dependence on parameters v_0, τ_V and $\dot{\varepsilon}$ the stress concentration on the notch face began to fall at a certain value of nominal stress, as a result of viscosity decrease in the region ahead of notch. When this takes place before the cohesion strength on the concentrator face is reached, there is no obstacle to further increase the nominal stress, and the material would eventually fail through intense shear. The opposite leads to cleavage.

The work mentioned above does not consider the low-temperature embrittlement in metallic glasses; however, we can make at least a qualitiative estimate of the occurrence of this phenomenon in dependence on $\dot{\varepsilon}$, v_0, τ_V and T. With decreasing temperature the rate of plastic deformation will also decrease, in agreement with the behaviour of the first term in Equation 1. The influence of the third term may be neglected considering the estimated value of parameters ΔG^m and Ω [10]. The values of v_0 and τ_V then determine if a material retains its plastic properties down to low temperatures at a given deformation rate. The initial concentration of free volume and the rate at which the amorphous structure is able to produce defects as a response to the stress concentration, are therefore the two parameters which decide if, at a given deformation rate, the low-temperature embrittlement takes place.

It follows from this rough consideration that Ni–Si–B glasses show either a higher initial density of deformation defects or that their structure is able to respond more quickly (by generating such defects) to the stress changes. Moreover, it is expected [5], that the non-isomorphism of the Ni_3B and Ni_3Si amorphous clusters in Ni–Si–B-type glasses will, itself, because of different thermal expansion coefficients, generate defects with high shear components or internal stresses, which would increase the ability of the amorphous structure to undergo an inhomogeneous shear. The model presented by Steif [16] does not take this factor into account, but it could be the main reason why plasticity is retained down to very low temperatures.

5. Conclusions

Measurements of the fracture toughness of metallic glasses on specimens with a centrally located crack at low temperatures reveal the following facts:

1. Ni–Si–B metallic glasses retain their plastic properties (as observed at room temperature) down to liquid helium temperature.

2. On the other hand, $Fe_{40}Ni_{40}B_{20}$ shows a tendency to embrittlement, this being dependent on the degree of structural relaxation.

The embrittlement of transition metal-based metallic glasses is manifested by a change in failure mechanism from a ductile failure in the plane of maximum shear stresses through the ductile failure in the plane of maximum normal stresses up to cleavage.

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