

Internal friction study of vacancy hardening in B2 FeAl alloys

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Abstract. Internal friction behaviour of B2 FeAl alloys has been examined to reveal the correlation of the microplasticity and thermal vacancies. The internal friction peak for Fe₆₀Al₄₀ appears at around 550 K, and the peak height increases with increasing quenching temperature. The curves of internal friction against the strain amplitude shift to larger strain amplitude as the quenching temperature increases. Analysis of the amplitude-dependent internal friction provides the plastic strain of the order of 10^{-9} as a function of effective stress on dislocation motion. It is found that the microflow stress at the plastic strain of 1×10^{-9} increases linearly with the square root of the net peak height. Remarkably, the microflow stress decreases with rising temperature but turns to increase above 500 K when measured after holding for 1 h at test temperatures. The anomalous increase in the microflow stress is caused by the creation of thermal vacancies at intermediate temperatures.

Introduction

Iron aluminide with B2 structure is one of the candidates for high-temperature structural materials because of its relatively high melting point, good oxidation resistance, high strength, low density and low cost. It has been well recognized that the physical and mechanical properties are strongly affected by atomic defects such as vacancies and antisite atoms. Major topics on the mechanical properties of B2 FeAl alloys are a drastic change in microhardness with the vacancy concentration [1] and an anomalous increase in yield stress with rising temperature [2]. Vacancy hardening can be recognized by noting thermal vacancies retained during quenching and/or slow cooling [1]. While many studies have observed peculiar dislocation behaviour such as dislocation climb [3], dislocation decomposition or reactions [4], it has been suggested that the yield stress anomaly is caused by solution hardening by an increasing number of immobile thermal vacancies [5]. Over the years, there has been disagreement on which is the most important mechanism controlling the yield stress anomaly [6].

Understanding the behaviour of atomic defects in B2 FeAl alloys is therefore extremely important to improve the performance at high temperatures. Internal friction technique is one of the best ways to reveal the defect properties. Schaefer *et al.* [7] observed an internal friction peak at 680 K for the stoichiometric FeAl, suggesting that it results from the reorientation of Fe antisite atoms (Fe_{Al}) and Fe vacancies (V_{Fe}) pairs. Golovin *et al.* [8] found an internal friction peak at about 450 K for Fe₃Al alloys and attributed it to a Snoek peak induced by the diffusion of interstitial carbon atoms under stress. More recently Wu *et al.* [9,10] have systematically investigated internal friction peaks over the wide range of B2 FeAl alloys. In addition to the temperature dependence of internal friction, the study of the amplitude-dependent internal friction is the most promising way to investigate the mechanical properties in the microplastic range [11]. In this study, internal friction behaviour of B2 FeAl alloys has been examined to clarify the correlation of the microplasticity and thermal vacancies.

Experimental

The FeAl alloys with Al content of 40, 45 and 53 at.% were prepared by repeating arc-melting of appropriate mixtures of 99.99% pure Fe and Al in an argon atmosphere. The ingots were homogenized at 1273 K for 48 h in vacuum. Specimens for internal friction measurements were cut

from the ingots with a SiC-blade saw to the size of $90 \times 10 \times 0.7 \text{ mm}^3$. Each sample was annealed at 1273 K for 1 h and quenched into water from various temperatures: quenching temperatures and hold times were 1273 K (for 1 h), 973 K (96 h) and 673 K (168 h). In addition, some samples were also furnace-cooled after heat treating at 673 K for 168 h to ensure a low initial vacancy concentration. X-ray diffraction measurements with monochromated Cu $K\alpha$ radiation detected only the B2 structure.

Internal friction was measured in vacuum by means of a free-decay method of flexural vibration with both ends of the sample free and at a frequency around 460 Hz in the fundamental resonant mode. After steady-state vibration for more than 60 s, the driving signal was turned off and a free-decay curve measured using a high-speed level recorder, where the logarithm of the vibration amplitude is recorded against time. As a measure of internal friction, the logarithmic decrement δ was determined from the slope of a tangent to the smooth envelope of the free-decay curve as a function of the maximum strain amplitude. The temperature dependence of internal friction was measured between 300 K and 673 K with a rising rate of 0.02 K/s.

Results and Discussion

Temperature Dependence of Internal Friction. Fig. 1 shows the internal friction δ in the furnace-cooled (closed symbols) and quenched (open symbols) samples of $\text{Fe}_{60}\text{Al}_{40}$ as a function of temperature. The internal friction peak can be found at around 550 K not only for the quenched samples but also for the furnace-cooled sample. The peak height significantly increased with increasing quenching temperature in the range from 673 K to 1273 K, so that the peak should be directly related to quenched-in defects. Wu *et al.* [9] previously observed a similar peak for $\text{Fe}_{57}\text{Al}_{43}$ at around 480 K using an inverted torsion pendulum and, according to their study, the large increase in the background above 600 K is due to the appearance of another internal friction peak.

It was also found that the internal friction peak for the furnace-cooled sample is dependent upon the measuring frequency f , shifting to higher temperature with increasing frequency. This indicates that the peak shows relaxational nature and thus should be associated with thermally activated relaxation process. The relaxation time τ and activation energy are thus expected to follow the Arrhenius relation:

$$\tau = \tau_0 \exp(H/kT), \quad (1)$$

where τ_0 is the pre-exponential factor, H the activation energy, k the Boltzmann constant and T the absolute temperature. At the peak position, $\omega\tau_p = 1$ should be satisfied, where $\omega = 2\pi f$ is the angular frequency and τ_p is the relaxation time at the peak temperature. Based on the Arrhenius plots of the peak temperatures and corresponding angular frequency, we obtained the activation energy $H = 1.08 \text{ eV}$, which is very close to that reported by Wu *et al.* [9]. They proposed that the internal friction peak is attributed to the reorientation of divacancies composed of Fe vacancies (V_{Fe}) and the nearest-neighbour Al vacancies (V_{Al}) [9].

Fig. 2 shows the internal friction δ in the furnace-cooled $\text{Fe}_{60}\text{Al}_{40}$, $\text{Fe}_{55}\text{Al}_{45}$ and $\text{Fe}_{47}\text{Al}_{53}$ as a function of temperature. The internal friction peak can be found at around 550 K for

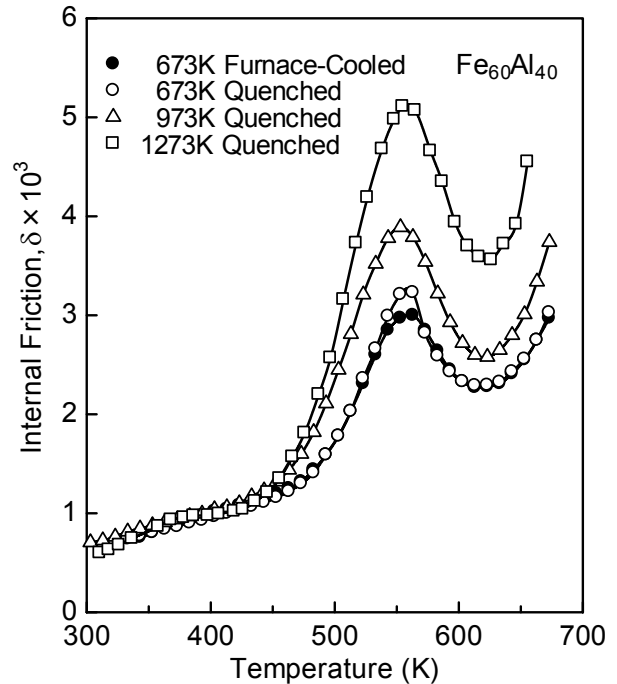
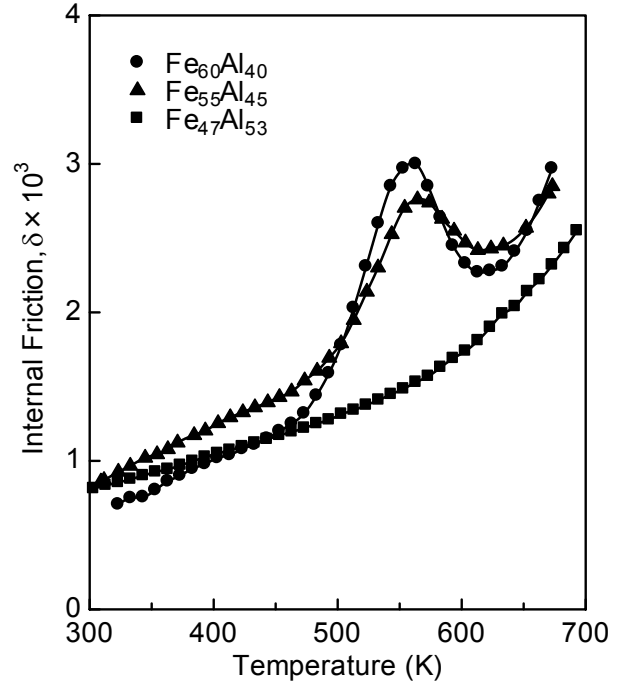


Fig. 1 Internal friction δ as a function of temperature for $\text{Fe}_{60}\text{Al}_{40}$ furnace-cooled and quenched from various temperatures.

both $\text{Fe}_{60}\text{Al}_{40}$ and $\text{Fe}_{55}\text{Al}_{45}$, but the peak height appears to decrease as the Al content increases. In case the composition of Al atoms exceeds the stoichiometric composition (50 at.%Al) for the B2 structure, excessive Al atoms will form Al antisite atoms (Al_{Fe}), giving rise to the disappearance of V_{Al} . In fact, the internal friction peak at around 550 K does not appear in the Al-rich alloy $\text{Fe}_{47}\text{Al}_{53}$, as shown in Fig. 2, which is consistent with the previous study [9]. Thus the present result is in line with a scenario that the internal friction peak originates from the reorientation of the divacancies composed of V_{Fe} and V_{Al} .

Fig. 2 Internal friction δ as a function of temperature for furnace-cooled $\text{Fe}_{60}\text{Al}_{40}$, $\text{Fe}_{55}\text{Al}_{45}$ and $\text{Fe}_{47}\text{Al}_{53}$.



Amplitude Dependence of Internal Friction. Fig. 3 shows the internal friction δ plotted as a function of the maximum strain amplitude ε_{max} measured at 300 K in the furnace-cooled (closed symbols) and quenched (open symbols) samples of $\text{Fe}_{60}\text{Al}_{40}$. It should be noted here that the curves of the amplitude dependence were reproducible when successively measured under the same conditions. Such reproducibility suggests that multiplication of dislocation does not occur during the measurements. The magnitude of the amplitude-independent part does not change systematically due to quenching. However, as compared with the furnace-cooled sample, the internal friction in the quenched samples becomes insensitive against the strain amplitude, so that the amplitude-dependent part appears at a higher strain amplitude as the quenching temperature increases.

According to the microplasticity theory [11,12], we can evaluate the microplastic stress-strain responses from the amplitude-dependent internal friction. For the data analysis, the $\delta(\varepsilon_{\text{max}})$ curves in Fig. 3 are approximated by a power function of ε_{max} ,

$$\delta(\varepsilon_{\text{max}}) = A\varepsilon_{\text{max}}^n + B, \quad (2)$$

where A , B and n are fitting parameters. Then the amplitude-dependent internal friction is converted to the plastic strain ε_p as a function of the effective stress σ :

$$\varepsilon_p(\sigma) = \frac{A(n+2)}{2^{n+2}} K(n) \left(\frac{\sigma}{E}\right)^{n+1}, \quad (3)$$

where E is the Young's modulus. $K(n)$ is the correction factor for strain distribution and is given for the flexural vibration as follows [13]:

$$K(n) = \frac{\sqrt{\pi} n + 3}{2} \frac{n + 3}{3} \Gamma\left(\frac{n + 4}{2}\right) / \Gamma\left(\frac{n + 3}{2}\right) \quad (4)$$

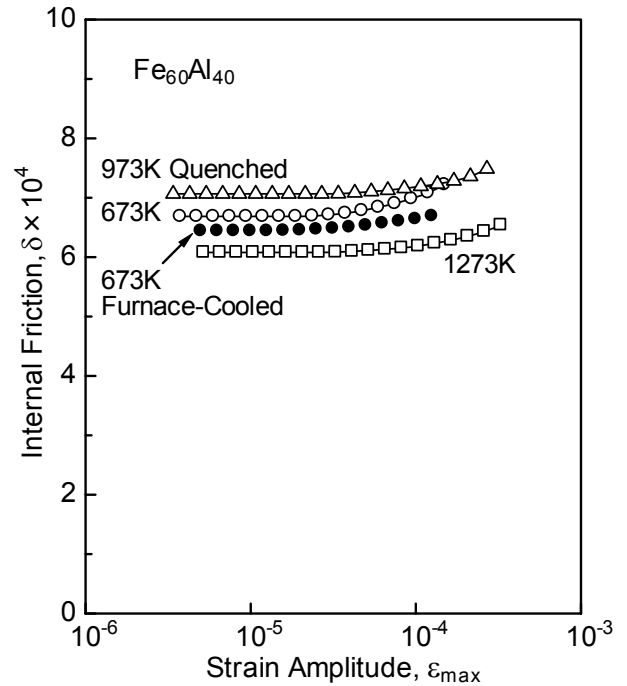
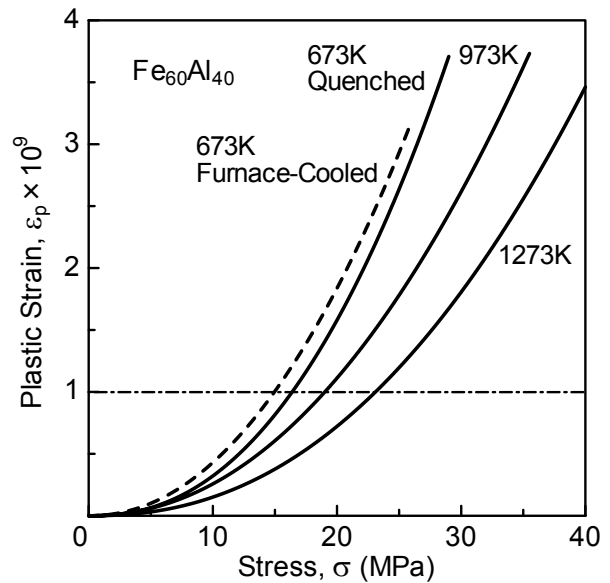


Fig. 3 Internal friction δ as a function of strain amplitude ε_{max} for $\text{Fe}_{60}\text{Al}_{40}$ furnace-cooled and quenched from various temperatures.

where Γ is the gamma function. Eq. 3 is applicable to evaluating the microplastic stress–strain responses from the internal friction data expressed by Eq. 2.

Fig. 4 shows the relation between the plastic strain ε_p and the effective stress σ for the furnace-cooled and quenched samples of $\text{Fe}_{60}\text{Al}_{40}$, corresponding to the stress-strain curves in the microplastic range: the curves are obtained from the results in Fig. 3. Although the value of ε_p is only of the order of 10^{-9} and as low as 0.01 % of the total strain, microplastic flow indeed occurs and the plastic strain increases nonlinearly with increasing stress. The curves tend to shift to a higher stress as the quenching temperature increases, thus indicating the suppression of microplastic flow due to quenching.

Fig. 4 Plastic strain ε_p as a function of effective stress σ in $\text{Fe}_{60}\text{Al}_{40}$ furnace-cooled and quenched from various temperatures.



Relation between Microflow Stress and Internal Friction. In order to clarify the vacancy hardening, we define the microflow stress at a constant level of the plastic strain, typically $\varepsilon_p = 1 \times 10^{-9}$ in Fig. 4. The microflow stress thus obtained can be regarded as a measure of the resistance to dislocation motion in the microplastic deformation. In Fig. 5, the microflow stress σ for the furnace-cooled and quenched samples of $\text{Fe}_{60}\text{Al}_{40}$ is plotted against the net peak height δ_{\max} of the respective internal friction peak shown in Fig. 1. The net peak height was obtained by subtracting the background of internal friction extrapolated linearly from the lower temperature region below 460 K in Fig. 1. It can be found that the microflow stress increases linearly with the square root of the net peak height, which is reminiscent of the relationship between the microhardness and the vacancy concentration [1]: the linear relationship would be expected from the solid solution strengthening model [14].

The vacancy concentration dependence thus qualitatively agrees with the variation in the microhardness, although the respective stress levels are quite different from each other. This qualitative agreement is surprising if one remembers that microplasticity is controlled only by dislocation motion under steady-state vibration, without accompanying strain hardening as in macroplasticity. Since the internal friction peak originates from the reorientation of the divacancies composed of V_{Fe} and V_{Al} , the net peak height should be proportional to the concentration of the divacancies, in contrast to the microhardness which is related to the vacancy concentration calculated from the thermodynamic model.

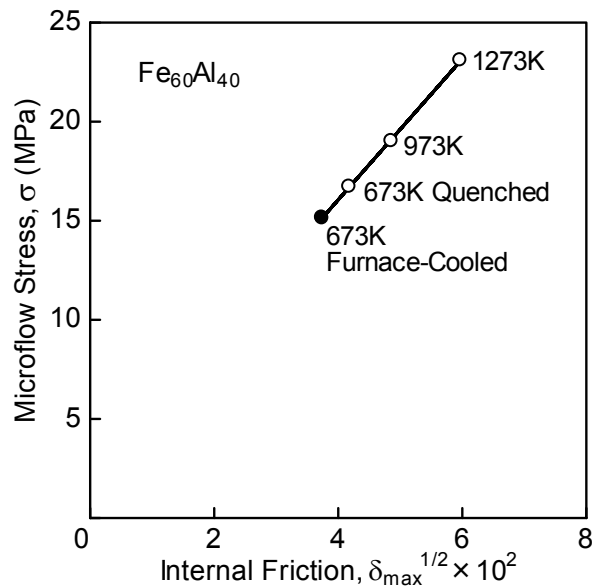


Fig. 5 Microflow stress σ plotted against the square root of the net peak height δ_{\max} of the internal friction peak in $\text{Fe}_{60}\text{Al}_{40}$ furnace-cooled and quenched from various temperatures.

Temperature Dependence of Microflow Stress. Because of a non-destructive nature of internal friction measurements, the test temperatures can be changed successively for a given sample. Firstly the amplitude-dependent internal friction was measured for the furnace-cooled $\text{Fe}_{60}\text{Al}_{40}$ as a function of temperature between 300 K and 673 K with a rising rate of 0.02 K/s. In the same manner as described above, we can evaluate the stress-strain responses in the microplastic range. In Fig. 6, the microflow stress σ at $\varepsilon_p=1\times 10^{-9}$ is plotted by the open circles as a function of temperature. It is seen that the microflow stress is almost constant during continuous heating, which is very different from the yield stress anomaly.

It should be noted here that new thermal vacancies are unavoidably generated when the samples are heated to elevated temperatures and brought to thermal equilibrium prior to testing [2]. Thus, the furnace-cooled sample was rapidly heated to 660 K in vacuum and held there for varying lengths of time, and then the amplitude-dependent internal friction was measured and analysed to evaluate the stress-strain responses. The change in the microflow stress σ at $\varepsilon_p=1\times 10^{-9}$ is plotted in Fig. 7 as a function of hold time at 660 K. Since the generation of the equilibrium vacancy concentration at high temperature is not instantaneous, the microflow stress just after heating to 660 K is found to be comparable to that obtained during continuous heating in Fig. 6. With increasing hold time at 660 K, the microflow stress is found to increase progressively presumably because the vacancy concentration increased. At hold times longer than 0.5 h, the microflow stress became independent of hold time. That is because the vacancy concentration reached the equilibrium value in about half an hour and there was no change in the vacancy concentration with further increases in hold time, being consistent with the behaviour of yield strength [2].

Thus the amplitude-dependent internal friction was measured after holding for 1 h at test temperatures between 300 K and 673 K. The change in the microflow stress σ is plotted by the closed circles in Fig. 6. Remarkably, the microflow stress first decreases with rising temperature but turns to increase above 500 K although the peak temperature for the microflow stress is still out of the temperature range examined. We believe that the vacancy concentration increases with increasing hold time, thus resulting in an increase in the microflow stress. Since the yield stress also starts to increase at around 500 K [2], we may speculate that the strength anomaly observed in both microplasticity and macroplasticity originates from the same mechanism although the stress levels are quite different from each other. It is not clear why the microflow stress first decreases with rising temperature up to 500 K, showing a lower stress than that obtained from continuous heating. We suppose that the vacancy concentration could become lower by low-temperature annealing than

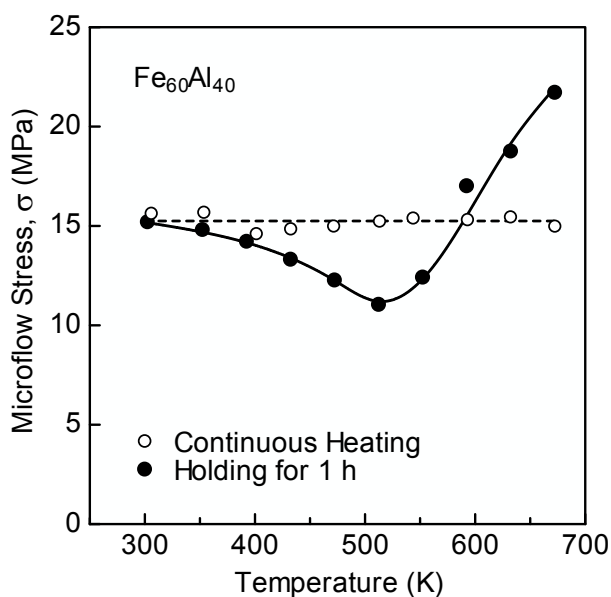


Fig. 6 Microflow stress σ as a function of temperature in $\text{Fe}_{60}\text{Al}_{40}$.

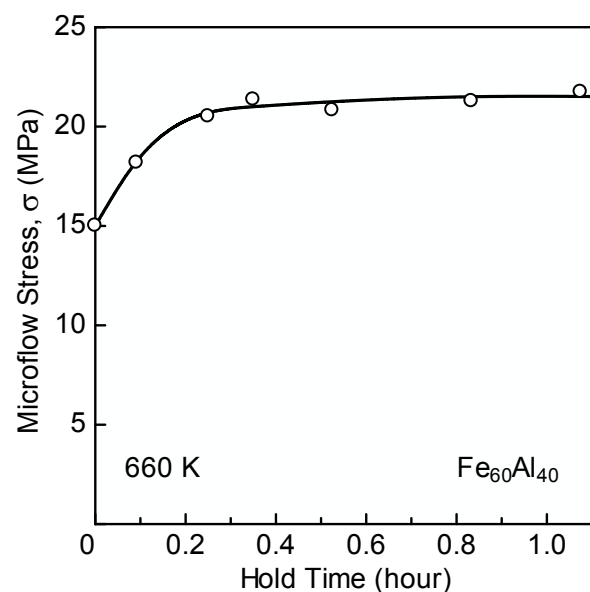


Fig. 7 Microflow stress σ as a function of hold time at 660 K in $\text{Fe}_{60}\text{Al}_{40}$.

that for the furnace-cooled sample. If the anomalous increase is due to dislocation mechanism i.e., climb dissociation [3] or glide decomposition [4], the microflow stress at high temperatures would be independent of how the sample arrived there. The vacancy mechanism, in contrast, depends sensitivity on the thermal history [2, 5], consistent with the result shown in Fig. 6. The present study strongly demonstrates that the anomalous increase in the microflow stress is due to hardening caused by the creation of thermal vacancies at intermediate temperatures.

Conclusions

Analysis of the amplitude-dependent internal friction in the furnace-cooled and quenched samples of Fe₆₀Al₄₀ provides the plastic strain of the order of 10⁻⁹ as a function of effective stress. The microflow stress increases with the quenching temperature and is proportional to the square root of the net peak height of the internal friction peak at around 550 K, which is reminiscent of the relationship between the microhardness and the vacancy concentration. The microflow stress is almost constant with temperature during continuous heating. When measured after holding for 1 h at test temperatures, however, the microflow stress first decreases with rising temperature but turns to increase above 500 K. The present study confirms that the strength anomaly in B2 FeAl alloys, as observed in both microplasticity and macroplasticity, could not be caused by thermally activated dislocation process but is most likely due to thermal vacancy formation at intermediate temperatures. The present internal friction data are very important in establishing the vacancy hardening model.

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