Fracture 1977, Volume 3, ICF4, Waterloo, Canada, June 19 - 24, 1977

FRACTURE AND TOUGHNESS OF BCC IRON ALLOYS AT CRYOGENIC TEMPERATURE

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INTRODUCTION

The structural steel commonly used for cryogenic service in liquid natural gas containers is the low carbon 9% Ni alloy. This steel has a relatively low toughness at 77 K and so should be rejected for use below 77 K. Since the application of superconducting technology and hydrogen as a clean energy source have become of major interest recently [1,2], new cryogenic structural materials for service at liquid helium temperature (4.2 K) or at liquid hydrogen temperature (20 K) are expected to be increasingly required in the future. A basic requirement is safety at such lower temperature. Consequently, high toughness is required above all. New cryogenic materials should possess both high strength at ambient temperature and toughness at service temperature [3]. Accordingly, this experiment was carried out to develop a new cryogenic iron alloy and to examine the fracture behavior based upon the understanding of the microscopic fracture mechanism of iron and iron alloy. The model presented is of relevance in explaining the effect of Ni on the brittle-ductile transition temperature of iron alloys.

CLEAVAGE FRACTURE OF IRON ALLOY

The property of low temperature brittleness is inherent in BCC metals. The dislocation theory has been applied through the various models [4] and although their models are suitable for understanding the grain size dependence of the fracture stress [5] and of the brittle-ductile transition temperature [6], the effect of alloying elements is not well explained. The following simple model could assist understanding the effect qualitatively.

I. Disclination model

The plastic deformation of BCC metals ascribes to the motion of screw dislocations at low temperature [7] but the fracture behavior might be assumed to be governed by the properties of edge dislocations [8]. Moreover, the cleavage fracture at low temperature always occurs at very small strain before yielding [9]. The long range pile-up of dislocations is unlikely to take place [10] but the properties in itself of the edge dislocation should be taken important account of. An edge dislocation in BCC lattice contains micro-crack just beneath its slip plane [11]. In addition, an edge dislocation can be easily transformed into a pair of wedge disclinations. The negative disclination might be assumed to be a micro-crack of the length of L, which is an important parameter associated with the Griffith condition of the micro-crack extension. The energy

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difference ΔE between an edge dislocation and a pair of wedge disclinations can be written as follows,

$$\Delta E = \frac{\mu b^2}{4\pi(1-\nu)} \ln \frac{2L}{r_o} + E_c - 2\gamma_s L \ , \qquad (1)$$

where μ is shear modulus, b Burgers vector, ν Poissons ratio, r_o the core radius of an edge dislocation ($\simeq$ b), E_c the core energy of an edge dislocation and γ_s the surface energy required to make new surfaces [12]. The detail of the calculation will appear elsewhere [13]. The stability of an edge dislocation in BCC lattice depends upon the core energy E_c, the length of the negative stable disclination, L and the surface energy γ_s. The above criterion suggests iron is a critical material at 0 K [13].

II. Effect of alloying elements

One of the important alloying elements reducing the transition temperature of iron alloys is Ni from the engineering viewpoint. Although the effect of Ni remains open to question, the summary of data can bring the conclusion that 12.5 at % Ni in α-Fe reduces the brittle-ductile transition temperature to 0 K [14]. The similar results are deduced for Pd and Pt. The value is simply calculated from the number of the first nearest neighbours of the core atom in BCC lattice. Consequently, the location of Ni group noble metal in the core site would decrease the core energy, followed by the stabilization of an edge dislocation in iron against micro-crack formation (that is transformation to disclination), whichever the effect is, elastic, electronic or magnetic. Another important element in iron alloy is carbon, of which contribution is not clear in carbide free concentration range. But the scavenging of free carbon is one of prominent techniques for lowering the transition temperature [15]. Actually, the increase of free carbon increases the grain size dependence k_y of Hall-Petch equation for yield stress with decreasing the test temperature. The effective cleavage fracture initiation stress σ_f associated with the effective surface energy γ_p, is given by Cottrell as follows,

$$\sigma_f = \frac{2\mu\gamma_p}{k_y} d^{-1/2} \ , \qquad (2)$$

where d is the average grain diameter [4]. The solute carbon affects the slip resistance through establishing the long range stress in BCC lattice [16]. Accordingly, the purification and the trapping free carbon of iron are a fairly effective method in improving the low temperature brittleness of iron and iron alloys [17].

APPLICATION

I. Development of high strength alloy for cryogenic services

The previous viewpoing leads us to the conclusion that a Fe-13.3% Ni (12.5 at %) alloy can not be fractured in a brittle manner even at 0 K. If the alloy is strengthened without embrittling the matrix, the combination of high strength and high toughness will be achieved in this simple system. One of the most prominent methods to increase the strength is supposed to be solid solution hardening mechanism. The addition of Mo, which is one of the most effective elements, also improves the resistance to temper embrittlement of Fe-Ni alloys [18]. The amount of Mo is determined within no presipitation range. Furthermore, since the grain size is another important factor controlling the toughness itself [19], the small addition of grain refinement, through the elimination of detrimental elements, is practically applicable technique. Ti is one of our choice, being affinitive to C, N, O and S. Table 1 gives the composition chosen for the experiment.

Total amounts of N, S and P of these alloys were below 80 ppm. Most of the alloys were quenched from the austenitizing temperature and little retained austenite (γ) was supposed to be present. Undetectable amount of austenite phase appears to play no important role in the mechanical behaviors.

II. Strength and toughness at cryogenic temperature

The stress-elongation curves of these alloys, which are as quenched, are shown in Figure 1, remarkably depending upon the test temperature. #3 alloy had relatively small grain diameter (prior austenite grain diameter of around 10 micron. The mechanism of plastic deformation changes at 4.2 K, so-called adiabatic deformation [20], since the twin was not observed in the tested specimens but the multiple necking appeared. Cleavage fracture was not fractographically observed on the surface broken even at 4.2 K. The grain size dependence of yield stress, k_y is much smaller than purified low carbon iron at every test temperature. #3 alloy yields 9.48×10^4 MP$\cdot$m$^{1/2}$ at 300 K, 1.01×10^5 MP$\cdot$m$^{1/2}$ at 77 K and 4.42×10^3 MP$\cdot$m$^{1/2}$ at 4.2 K, respectively. The smaller k_y is supposed to increase fairly the effective cleavage fracture stress in the equation (2). The toughness was represented by the impact energy obtained by V-notch Charpy impact test. The impact energy considerably depends upon the heat treatment carried out on these materials. The best values of these alloys at 77 K are shown in Table 2.

The yield stress are listed together. Prior to the final test at 4.2 K, the estimation of the toughness can be made by the aid of the correlation between the toughness and the reduction of area [21]. The most underestimated value of these alloys was about 60 J, which is supposed to be much higher than that obtained usually in cleavage fracture. Consequently, these experimentally arranged alloys give the high toughness even at cryogenic temperature, while higher strength is maintained at room temperature. Brittle behavior was not observed in every material even at 4.2 K.

CONCLUSION

1. Nickel, which is one of the most effective elements reducing the transition temperature, is supposed to decrease the core energy of an edge dislocation of BCC iron, followed by the more difficult transformation.
2. The simple consideration leads that Fe-13.3 % Ni alloy is the most optimum composition.
3. Experimentally made high strength-tough alloys for cryogenic use was tested at service temperature and cleavage fracture was not observed even at 4.2 K.
4. The recommended composition was Fe-13.3 % Ni - 3 % Mo - 0.2 % Ti.

ACKNOWLEDGEMENTS

The authors would like to thank the members of National Research Institute for Metals at the melting and rolling section, the work shop, the cryogenic section and the chemical analysis section for the present work. They also acknowledge Professor Dr. T. Araki, the director of NRIM for permission of the presentation.

REFERENCES

1. BIRMINGHAM, B. W. and SMITH, C. N., Cryogenics, 16, 1976, 59.
2. WILLIAMS, L. O., Cryogenics, 13, 1973, 693.
3. PARKER, E. R. and ZACKAY, V. F., Engineering Fracture Mech., 5, 1973, 147.
4. COTTRELL, A. H., Trans. AIME, 212, 1958, 192.
5. STROH, A. N., Proc. Roy. Soc., A223, 1954, 404.
6. PETCH, N. J., Phil. Mag., 3, 1958, 1089.
7. TAKEUCHI, T., Rev. Deform. Behav. Mater., 1, 1975, 119.
8. SMITH, E., Mater. Sci. Engg., 17, 1975, 125.
9. KNOTT, J. F., Fund. Fract. Mech., Butterworth, London, 1973.
10. ROSENFIELD, A. R. and Kanninen, M. F., Phil. Mag., 22, 1970, 143.
11. BULLOUGH, R. and Perrin, R. C., Dislocation Dynamics, McGraw-Hill, N.Y., 1968, 175.
12. LI, J. C. M., Surface Science, 31, 1972, 12.
13. ISHIKAWA, K., unpublished data.
14. ISHIKAWA, K. and TSUYA, K., Proc. 19th Japan Cong. Mater. Res., 1976, 57.
15. JOLLY, P., Met. Trans., 2, 1971, 341.
16. OWEN, W. S. and ROBERTS, M. J., Dislocation Dynamics, McGraw-Hill, N.Y., 1968, 357.
17. ZACKAY, V. F., PARKER, E. R., MORRIS, J. W., Jr. and THOMAS, G., Mater. Sci. Engg., 16, 1974, 201.
18. NAGASHIMA, S., OOKA, T., SEKINO, S., MIMURA, H., FUJISHIMA, T., YANO, S. and SAKURAI, H., Tetsu-to-Hagané, 58, 1972, 128.
19. YOKOTA, M. J., SASAKI, G. and HORWOOD, W. M., Mater. Sci. Engg., 19, 1975, 129.
20. BASINSKI, Z. S., Proc. Roy. Soc., A240, 1957, 229.
21. HANNERZ, N. E. and LOWERY, F. J., Met. Const., 7, 1975, 21.

Table 1 Composition of used materials (wt%)

Materials	Ni	Mo	Ti	C	Fe
#1	13.3	3.0	0.10	0.004	Bal.
#2	13.3	3.0	0.20	0.004	Bal.
#3	13.3	3.0	0.40	0.004	Bal.
#4	13.3	3.0	0.80	0.004	Bal.

Table 2 Maximum toughness of these alloys (J) and corresponding yield stress at 300 K (MPa)

Materials	#1	#2	#3	#4
$_vE_{77}$	248	199	162	61.8
Yield stress	616	759	624	834

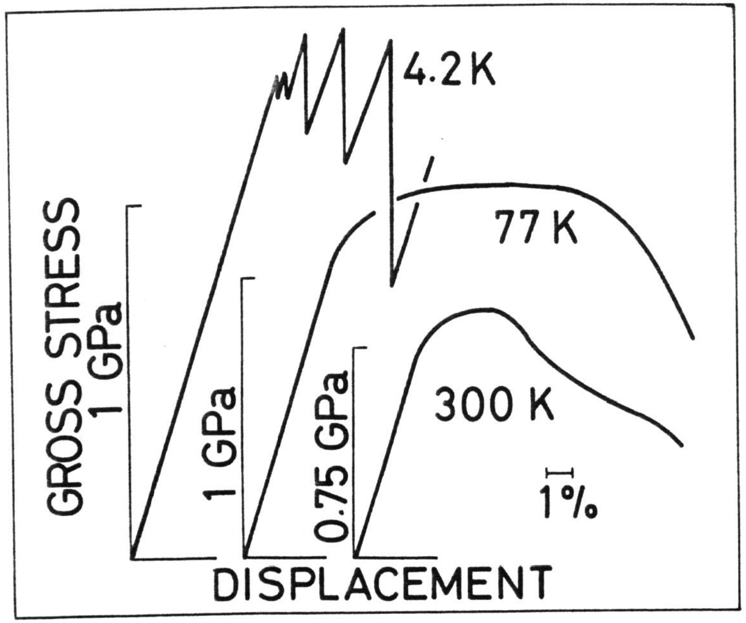

Figure 1 Stress-elongation curves of #3 alloy at 4.2, 77 and 300 K, respectively