Cleavage-like Fracture at Low Temperatures in an 18Mn-18Cr-0.5N Austenitic Steel

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(Received on December 1, 1989; accepted in the final form on January 26, 1990)

The low temperature embrittlement in an 18Mn-18Cr-0.5N retaining ring steel has been investigated by means of tension and Charpy impact tests. The effects of grain size, strain rate and pre-deformation were examined.

The ductile to brittle transition (DBT) behaviour associated with the cleavage-like fracture has been found, where the fractured plane was regarded as {111}. The traces of intense planar plastic flow which may be related with twinning, slip band, or epsilon martensite were observed on the surface of plastically deformed specimen. When the pre-deformation was applied to the specimen at room temperature, the fracture stress at 77 K was slightly increased but the fracture mode was still cleavage-like. The DBT temperature (DBTT) is scarcely influenced by grain size, although the fractured surface unit is considered as austenite grain size. This is discussed based on the experimental data on the grain size dependences of flow stress and fracture stress. When the strain rate in tension test is increased up to 2.7×10^{-1} /s, the fracture mode at 77 K has been found to change from cleavage-like to dimple. In this case, the plastic deformation occurs very locally and heavily, so as to induce the necking of the specimen.

KEY WORDS: high Mn steel; retaining ring steel; low temperature embrittlement; cleavage; grain size; cooling rate.

1. Introduction

It is very unusual to occur the transgranular brittle fracture (cleavage) at low temperatures in face-centered cubic (fcc) alloys. It is also predicted from theoretical approach¹⁾ that the fcc metals and alloys cannot cleave easily. The cleavage-like fracture at cryogenic temperatures in high N containing Fe-Cr-Mn austenitic steels were firstly studied by Defilippi *et al.*,²⁾ but scarcely discussed after them.³⁾ In recent years, as high Mn austenitic steels have been developed according to the demand and development of cryogenic engineering, attention has again been paid on this brittle fracture.⁴⁻¹¹⁾ Tobler and Meyn⁶⁾ have found this kind of austenite {111} transgranular fracture in an Fe-18Cr-3Ni-13Mn-0.37N steel at liquid helium temperature by fracture toughness test. Following the ASTM definition on terminology (ASTM's standard E616-82¹²⁾), this fracture could be called as cleavage. However, they called it as cleavage-like, because they could not observe the river pattern on the flat facet, but, instead of it, recognized the fine slip traces at higher magnification by scanning electron microscopy (SEM) observation. That is, this fracture mechanism was considered as slip-band cracking (not pure cleavage).⁶⁾ Since their study was performed under special conditions, much has been remained unclear. Because the solid solution hardening by nitrogen plays a very important role in designing high strength nonmagnetic steels, simpler examination to make clear the detailed fracture mechanism must be carried out. This paper shows several

new findings on this unusual brittle fracture, by means of tension and impact tests. Concerning the terminology, it might be questionable whether 'cleavage-like' is proper or not. However, since the fracture mechanism is not clarified enough, the term 'cleavage-like' by Tobler and Meyn is used in this paper to avoid confusion.

2. Experimental Procedure

The material used in this experiment was one of the retaining ring steels which was received as hot forged and solution-treated (at 1323 K for 3.6 ks, water-quenched) condition from The Japan Steel Works Ltd. The chemical compositions of the material are 0.070 C, 0.30 Si, 18.82 Mn, 0.017 P, 0.003 S, 0.20 Ni, 17.22 Cr, 0.05 Mo, 0.12 Cu, 0.008 Al, and 0.513 N (all in wt%). Some specimens were machined from as received bars with a cross section of 30 by 30 mm. After hot- and cold-rollings, the tension and Charpy impact test specimens were machined. The tension test specimen is a plate one with 1.5 mm thickness, 3 mm width and 30 mm gage length. The three kinds of Charpy impact test specimens were made, *i.e.*, (a) ASTM standard V notch specimen, (b) full size specimen with a sharp notch (radius at a tip of notch was 0.165 mm), and (c) half size one (thickness of the specimen was half of (b)). The heat treatments given to specimens in Ar gas atmosphere and resultant austenite grain size by the relevant heat treatments (determined by ASTM method) are listed in Table 1. The Charpy impact tests were

Table 1. Solution treatment conditions and resultant grain size.

Heat	Grain size (μm)
1 473 K, 86.4 ks	576
1 473 K, 3.6 ks	156
1 373 K, 3.6 ks	49
1 273 K, 3.6 ks	26

carried out at various temperatures which were adjusted by using isopentane and liquid nitrogen. The tension tests were conducted on a gear-driven type machine at an initial strain rates of 2.7×10^{-3} , 2.7×10^{-2} , and 2.7×10^{-1} /s (cross head speeds of 5, 50, and 500 mm/min), at 77, 163, and 293 K, respectively. Fractography was examined by using a scanning electron microscope (Shimadzu EMX-SM).

3. Results and Discussion

3.1. DBT Behaviour and the Features of Fracture at 77 K

The data obtained by the specimens solutionized at 1 373 K for 3.6 ks will be used in this section if not specially commented. Fig. 1 shows the DBT behaviour in this alloy by standard ASTM specimens. Here, next points must be noted: one is a very clear DBTT as can be seen in ferritic steels and another is a clear upper shelf that exists just before the DBTT. As usual, the toughness of a highly strengthened fcc alloy decreases gradually with a decrease in temperature (For instance, see the dashed lines in Fig. 1 which was obtained in another kind of retaining ring steel, *i.e.*, Fe-18Mn-5Cr-0.5C steel⁵⁾). To examine the characteristics in plastic flow, the tension tests were performed and the results are shown in Fig. 2. A very strong temperature dependence of 0.2 % proof stress is a common feature in the austenitic steels containing large amount of nitrogen. For comparison, the trends of 0.2 % proof stress *vs.* temperature relations in some austenitic steels are presented from the literature.¹³⁾ An increase in strength by lowering the test temperature is outstanding in N33, 205, and the present steel (all high N containing steels) compared with those in SUS304, SUS316, and A286 (hardened by precipitation).

On the other hand, the elongation (EL) and reduction in area (RA) are deteriorated with lowering the test temperature. The very poor ductility and toughness at 77 K are related with the occurrence of transgranular brittle fracture.

Fractographically, the fractured surface at 77 K either by tension or impact, consists of very flat facets as shown in Fig. 3. When the fractured surface was protected by Ni-plating, sectioned, polished, and etched, the aspect like in Fig. 4 could be obtained. The dense planar lines seemed to be either epsilon martensite (ϵ), twinning, or slip band. The evidence of ϵ martensite was not obtained by the X-ray analysis. Although, the present authors have not succeeded TEM (transmission electron microscopy) analysis on the identification of this product, Tanaka *et al.*⁹⁾ have exhibited that most of them were analysed as ϵ

Fig. 1. Absorbed energy (Charpy impact value) as a function of test temperature.

TS: Tensile strength PS: 0.2 % proof stress
RA: Reduction in area EL: Total elongation
(Initial strain rate: 2.7×10^{-3} /s)
----: 0.2 % proof stress drawn for comparison
(after Ref. 13)

Fig. 2. Relationship between tensile properties and test temperature.

Fig. 3. Typical fractography of fractured surface at 77 K by impact test.

martensite in this kind of retaining ring steel. They have reported the diffraction spots to indicate twinning were rarely found.

Fig. 4. Aspect of the cross-section of fractured specimen at 77 K by impact test, where planar traces appeared after Ni-plating, polishing and etching (etchant: $\text{HCl} + \text{HNO}_3$).

Since all of these possible products by plastic deformation are known to occur on $\{111\}$ plane, the traces of $\{111\}$ could be drawn like in Fig. 4 which suggested the fractured plane was almost parallel to one of $\{111\}$. From the two-planes analysis, the fractured plane was also considered as $\{111\}$. Concerning the plane of this kind of fractured surface, Shibata and Fujita have obtained an evidence of $\{111\}$ cleavage by the etch-pit technique in an Fe-32Mn-7.5Cr-0.32N steel.⁴⁾ Tobler and Meyn have also determined as $\{111\}$ by Laue X-ray diffraction in an Fe-18Cr-3Ni-13Mn-0.37N steel.⁶⁾ They examined the fractured surface obtained by fracture toughness test at 4.2 K in detail and their opinion on this fracture is not pure cleavage but cleavage-like fracture which means slip-band cracking, as described in Introduction. In fact, the river pattern was hardly observed in this study, either. Instead, lots of fine straight relieves were recognized on the flat facet (for instance, see Figs. 3, 8, and 12). On the other hand, Fig. 5 shows the initiation and propagation of a sub-crack in a tension test specimen (the grain size was $156 \mu\text{m}$). The crack is estimated to be initiated from the side surface of the specimen. A lot of surface relieves produced by tensile deformation can be seen and all of them are considered to show $\{111\}$ traces as discussed above. Therefore, if one can identify the four different $\{111\}$ traces, the crystal orientation of a grain can be determined.¹⁴⁾ Using this method, the orientations of grains A and B in Fig. 5(a) were estimated as shown in (b). Here, the cleaved plane is predicted roughly as $(1\bar{1}\bar{1})$ in grains A and B. Although the crack in grain A is estimated almost perpendicular to the tensile axis $[6\ 5\ 10]$, it looks to be declined in grain B. Thus, it must be better to consider the normal cleavage occurred in grain A. However, the result in grain B might be in a good agreement with Tobler and Meyn's opinion of slip-band cracking. Looking at the fractured surface by SEM,

some facets were declined sharply against the macroscopic fractured surface: this suggested the cleaved plane was an active slip plane, but some were almost parallel (non-active slip plane). Therefore, it is possible to consider that the fracture mechanism of this steel changes depending on a given stress condition.

The present authors have observed this kind of brittle fracture takes place in more stable austenitic steels,¹⁰⁾ so that the role of ϵ martensite is not concluded as an essential factor. One more suspecting point for the origin of this transgranular cracking is a role of mechanical twinning as is well known in ferritic steel.¹⁵⁾ The surface relief in Fig. 5 and aspect in Fig. 4 look similar to the twinning. Here, the pre-deformation was given at room temperature in order to change the deformation manner at 77 K. As a result, the fracture stress was slightly increased by pre-deformation as shown in Fig. 6, but the fracture mode was not changed. The TEM results by Tanaka *et al.*⁹⁾ were quite negative for the role of twinning against this fracture. These results look to conflict a little with the earlier reports.^{2,3)} As different aspects have been obtained, more systematic experimental examination must be required. Especially, it must be important to examine the crack initiation mechanism being classified from the crack propagation stage in the future.

3.2. Effect of Grain Size

The Charpy impact tests were carried out for specimens received various solid solution treatments, *i.e.*, with different grain sizes. When solutionizing temperature is too low, the nitride precipitation occurs. On the other hand, if solutionizing temperature is higher than 1473 K, the intrusion of δ ferrite would become problem. The data used here were considered as austenitic single phase by X-ray diffractometer and optical microscopy.

Fig. 7 shows the results of Charpy impact tests.

- (a) Optical micrograph
- (b) Result of trace analysis in grains A and B. In a grain C, 4 kinds of (111) traces could not be obtained.

Fig. 5.
An example of crack initiation and propagation observed on the specimen's surface by tension test at 77 K.

Fig. 6. Effect of pre-deformation at room temperature (tension) on flow curves at 77 K.

Very interesting is the fact that DBTT is not changed or rather increased with a decrease in grain size, being different from the case of ferritic steels. Similar results were reported by Kawai *et al.*⁷⁾ Fig. 8 presents the influence of grain size on fractography at 77 K. The fracture unit corresponding to the microstructure can be regarded as austenite grain size. Therefore, reducing the fracture unit associated with grain refining is concluded of no use to lower the DBTT. This is just a reverse trend compared with cases of ferritic steels.

An increase in DBTT with grain refining has been observed in several metals and alloys, for instance, in Mo,^{16,17)} Be,¹⁸⁾ Mg,¹⁸⁾ and an Al-Zn alloy.¹⁹⁾ Concerning the relation between DBTT and grain size, a following explanation has been successfully given. A schematic illustration of temperature dependences of the yield strength (0.2 % proof stress) and fracture stress are sketched in Fig. 9. Here, the DBTT is assumed to be defined from the comparison with yield

Fig. 7. The effect of grain size on Charpy impact energy as a function of test temperature.

strength and fracture stress, a cross point of two curves is regarded to be connected with DBTT. When the grain size dependences of the yield strength and fracture stress are approximated by well known Hall-Petch equations;

$$\sigma_y = \sigma_{0y} + k_y d^{-1/2} \dots\dots\dots(1)$$

$$\sigma_f = \sigma_{0f} + k_f d^{-1/2} \dots\dots\dots(2)$$

where, d : grain size (or grain diameter)

σ_f : fracture stress

σ_y : yield strength

 $\sigma_{0y}, \sigma_{0f}, k_y, k_f$: constants

as is often done, the grain size dependence of DBTT can be discussed from a comparison of k_y and k_f . That is, with increasing of grain size, DBTT would be predicted to increase if $k_y > k_f$, or decrease if $k_y < k_f$.

 (a) 576 μm (b) 49 μm (c) 26 μm

Fig. 8. Change in fractography with grain size.

The martensitic steel (for example 12 Ni steel²⁰⁾), must be a good example of the latter case ($k_y < k_f$). The fracture unit of the lath-martensitic steel has been considered as either austenite grain, packet, or block, depending on the microstructural characteristics,²¹⁾ all of which are expected to decrease by grain refining. Therefore, it might be considered the fracture stress is increased by grain refining, but the yield strength is not influenced much by grain size.²⁰⁾ In such cases, DBTT could be expected to lower with a decrease in grain size. In a bcc iron, the k_f (both for cleavage and intergranular fracture) has been reported four to five times larger than k_y (for lower yield point).²²⁾

Fig. 9. Schematic illustration to explain the change in DBTT by grain refining.

Point A is for larger grain size and points B to D can be assumed for smaller grain size depending on relative change in flow stress and fractured stress.

However, if the grain size dependence of fracture stress is lower (see curve ① in Fig. 9), DBTT would increase by grain refining. In cases of some hcp metals¹⁸⁾ and an Al-Zn alloy¹⁹⁾, k_y was reported larger than k_f (for instance, 12.1 (k_y) and 3.7 MPamm^{1/2} (k_f), respectively, in the Al-Zn alloy¹⁹⁾).

The influence of grain size on tensile properties was examined. The data on 0.2 % proof stress and fracture stress are plotted against test temperature in Fig. 10. As long as compared by a change in absolute value, the grain size dependence in fracture stress (true fracture stress was estimated by using the reduction in area, then it remains some problem on accuracy) is found higher than that in 0.2 % proof stress, so that it seems to contradict the explanation by Fig. 9.

However, the values of k_y : 27 (at 293 K), 43 (at 163 K), and 52 MPamm^{1/2} (at 77 K) in this steel should be noted, because the value of k_y in mild steels is around 20 MPamm^{1/2} and changes little by test temperature.^{22,23)} As Shibata pointed in his review,¹¹⁾ the understanding in the value of k_y in austenitic steels is in confusion: various different results have been reported, although k_y in an austenitic steel has been considered almost half of that in a ferritic steel in a textbook.²⁴⁾ The values of k_y in an Fe-Cr-Mn steel, reported by Yamamoto *et al.*,²⁵⁾ were approximately 25 (*RT*) and 50 MPamm^{1/2} (4 K), which are consistent with the present data. Furthermore, the k_y in Fe-Cr-Mn steels containing 0.5 to 0.8 wt% N were estimated between 50 and 60 MPamm^{1/2} at 77 K.¹⁰⁾ Then, the large k_y at low temperatures seems to be one of the intrinsic features in N containing Fe-Cr-Mn steels.

A change in k_y with temperature was found also in Mo (60 MPamm^{1/2} at 195 K while 38 MPamm^{1/2} at 293 K). Since the large temperature dependence of k_y in Mo is believed due to operating slip systems particular to this crystal structure, the deformation mechanism associated with thermal activation process in this steel must yield this unusual trend in k_y .

Therefore, although quantitatively sufficient explanation cannot be obtained, the idea in Fig. 8 is believed basically very promising, thinking about the roughness in the estimation of fracture stress in Fig. 9. The unusual DBTT *vs.* grain size relation in this steel can be considered to be ascribed of the characteristic deformation mechanism.

3.3. Effect of Strain Rate on Deformation and Fracture

Fig. 11 shows nominal stress-strain curves at room temperature (*RT*: 293 K) and liquid nitrogen temperature (77 K). At *RT*, the flow stress is found to increase with an increase in strain rate, where the strain rate dependence seems to be a little higher than in other austenitic steels. This might be related with the extremely higher temperature dependence of strength in Fig. 2. The total elongation is decreased with increasing of strain rate. To be noted here is

Fig. 10. Influence of grain size on 0.2 % proof stress and fracture stress as a function of test temperature.

Fig. 11. Effect of strain rate on nominal stress-strain curves at room temperature and 77 K.

the results at 77 K. At the highest strain rate ($2.7 \times 10^{-1}/s$), the plastic flow was localized within a narrow band from very beginning of the deformation, and to induce necking. After that, the specimen fractured in a ductile manner (dimple), so that *RA* was about 60 % in spite of very small *EL*. The flow curve at this strain rate in Fig. 11 is abnormal one. The fractured surfaces are shown in Fig. 12. The cleavage-like fracture was observed at lower strain rate, while dimple was covered all over the fractured surface at the highest strain rate. When the surface of specimen was observed, slip lines were found only in the necked region. This is quite contrast with other

Strain rate: (a) $2.7 \times 10^{-3}/s$ (b) $2.7 \times 10^{-2}/s$ (c) $2.7 \times 10^{-1}/s$

Fig. 12. Influence of strain rate on fractography at 77 K (99 μm).

specimens tested at lower strain rates, where slip lines (or surface relieves) were observed almost uniformly through the parallel portion of the tension specimen. It suggests that a kind of work-softening takes place at higher strain rate at 77 K. It must be considered that temperature was increased very locally caused by plastic flow. However, this phenomenon has not been found in other steels (the serration at liquid helium temperature occurs in general,¹¹⁾ but the aspect is a little different from this case). The question why only this steel shows such unusual behaviour would be closely connected with solution strengthening by N. The clear answer has not been obtained against the question why N is so effective to enhance the low temperature strength (see Fig. 2). This strengthened microstructure by N is thought to be very fragile under some condition, for instance, cyclic loading.²⁶⁾ Although the detailed N strengthening mechanism is not clarified, one promising idea (hypothesis) has been proposed by Shibata and Fujita.^{26,27)} They studied about cyclic deformation behaviour and thought that the fatigue softening occurred through a breakdown of special solid solution hardening due to IS (interstitial and substitutional atoms) complexes during cyclic deformation. If this kind of IS complexes is proved, a local breakdown of resistance against plastic flow could happen being accompanied with the local temperature rise. These unusual strain rate dependences have been recognized in other N strengthened Fe-Cr-Mn alloys¹⁰⁾ which will be reported by another paper where the influence of stress condition (specimen's geometry) and the difference between tension test and impact test by which cleavage-like fracture occurred even at very high strain rate will be discussed.

4. Conclusions

The main findings obtained in this investigation would be summarized as follows.

(1) A transgranular fracture occurs in this steel along {111}. The fracture mechanism is not sufficiently clear because some results suggest pure cleavage but others cleavage-like, *i.e.*, slip-band cracking. The crack initiation is considered to be related with heterogeneous plastic flow before the cracking, but the detail in the coarse linear product is still unclear.

(2) The effect of grain size on ductile to brittle transition temperature (DBTT) is very small or rather increases by grain refining. This is considered to be related with unusual grain size dependence of the yield strength. The slope in Hall-Petch plotting is higher and strongly temperature dependent being quite contrast with ferritic steels.

(3) When the strain rate is increased up to 2.7×10^{-1} /s, the fracture mode is drastically changed from cleavage-like to dimple at 77 K. The particular microstructure strengthened by nitrogen is considered to induce the local softening in couple with local temperature rise.

Acknowledgements

Authors wish to appreciate The Japan Steel Works Ltd. for the supply of the experimental material. They thank Messrs. Y. Adachi and S. Ishino (students of Ibaraki University) for their experimental assistance. They are also grateful to Professor Shibata of The University of Tokyo for helpful discussion.

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