Toughness of Ultrafine Grained Ferritic Steels Fabricated by ARB and Annealing Process

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A plain IF steel and a P-added IF steel having various ultrafine grain sizes from 0.24 to 11 μm were fabricated by the accumulative roll bonding (ARB) process followed by annealing. Dynamic fracture toughness of the ultrafine grained IF steels was investigated as a function of grain size by miniaturized Charpy impact test. The static strength of the IF steels significantly increased with decreasing the grain size, while the uniform elongation was limited in the ultrafine grained samples. A number of delamination appeared in the impact-tested specimens, especially in the ultrafine grained materials at low temperatures. It was concluded that the frequent delamination is not owing to insufficient roll-bonding in the ARB specimens but it is rather a characteristic feature of the ultrafine grained materials fabricated through heavy deformation. Because of the delamination, the absorbed energy in the impact test continuously decreased with decreasing the test temperature. On the other hand, an obvious change from the ductile fracture surface characterized by dimples into the brittle fracture surface mainly due to intergranular fracture was recognized at a certain low temperature. The ductile-brittle transition temperature determined from the microscopic fracture surfaces greatly decreased with decreasing the grain size, and finally no brittle fracture happened even at -190°C when the grain size was smaller than 5 μm or 2 μm in the plain IF steel or the P-added IF steel, respectively. It was concluded that the ultra-grain refinement is quite effective to improve the low-temperature toughness of ferritic steels and that it is possible to make phosphorus substantially harmless by grain refinement.

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1. Introduction

Grain refinement has always been a key subject in the microstructure control of steels. At the moment, the limitation of grain refinement in the bulk structural steels is around 10 μm . The minimum grain size previously achieved in commercial steels is 5 μm for the plates fabricated through a TMCP (thermomechanically controlled processing) called controlled-rolling.¹⁾ On the other hand, the research works on ultra-grain refinement of steels have been energetically conducted in the last decade in order to drastically improve the mechanical properties.²⁾ Those studies have been trying to realize the mean grain size smaller than a few μm . One of the promising ways to fabricate ultrafine grained metallic materials having bulky dimensions is the severe plastic deformation (SPD), and various kinds of special SPD processes have been developed.³⁾ The present authors have also invented a SPD process named the accumulative roll bonding (ARB)³⁻⁶⁾ using rolling deformation and succeeded in fabricating the ultrafine grained (UFG) bulk metallic materials including various kinds of steels, whose mean grain sizes are smaller than 1 μm .^{3,6)}

In order to put the UFG materials in practical use, by the way, it is necessary to know their mechanical properties in details. Nevertheless, systematic data for the mechanical properties of the UFG materials are scarcely available. This lack is mainly due to the difficulty to obtain enough size and amount of the UFG samples through special TMCP or SPD techniques in laboratory scale. The ARB process, however, has an advantage to fabricate large bulky samples even in university laboratories,^{3,6)} though the shape of the obtained materials is basically a sheet. The ultrafine grain size

obtained by the ARB can be further changed by subsequent annealing,^{7,8)} so that bulky samples having various mean grain sizes ranging from 100 nm to 100 μm are prepared for systematic data collection. By the use of this advantage, Tsuji *et al.*^{9,10)} clarified the strength and ductility of the UFG aluminum and iron systematically as a function of their grain size. The purpose of the present study is to clarify the dynamic fracture toughness of the UFG ferritic steels as a function of the grain size. Although toughness is an important mechanical property of structural steels, there are again little data¹¹⁾ about toughness of the UFG steels. As a typical ferrite single-phase steel, an ultralow carbon interstitial free (IF) steel is used in this study. The ARB process followed by annealing is carried out in the present investigation to provide the bulky samples having various UFG sizes. Because the ARB can only fabricate sheet materials having limited thickness, the miniaturized Charpy impact test^{12,13)} is carried out to measure the dynamic fracture toughness of the materials.

Additionally, the present study focuses on another interest. It is well-known that some kinds of impurities lower the toughness of steels. This is because such impurities segregate at grain boundaries and enhance intergranular fracture.¹⁴⁾ In the UFG steels, however, there are so many grain boundaries²⁾ that the concentration of the impurities segregated at the grain boundaries is expected to be diluted even in the material having the same chemical composition. That is, it might be possible to decrease the bad influence of the impurities on toughness by ultra-grain refinement. In order to throw light on this possibility, a P-added IF steel is also used in the experiment. Phosphorus is a typical element to induce intergranular fracture through grain boundary segregation not only in martensitic steels¹⁵⁾ but also in ferritic steels.^{14,16)}

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Table 1 Chemical compositions of the IF steels studied (mass %).

	(wt.%)							
	C	Si	Mn	P	S	Ti	Al	Fe
plain IF	0.002	<0.01	0.13	<0.001	0.006	0.050	0.085	bal.
P-added IF	0.002	<0.01	0.14	0.041	0.006	0.050	0.069	bal.

2. Experimental

Two kinds of Ti-added ultralow carbon IF steels whose chemical compositions are shown in Table 1 were used. One is a plain IF steel and the other is a 0.041%P-added IF steel. As-received sheets cold-rolled by 80% were annealed at 800°C for 1.8 ks to complete recrystallization. The materials showed fully recrystallized grains with equiaxed shapes, and the mean grain sizes were 15 μm and 12 μm for the plain IF steel and the P-added IF steel, respectively. The fully annealed sheets were provided to the ARB process. Two pieces of the sheets 1 mm thick, 30 mm wide and 250 mm long were stacked after degreasing surface by acetone followed by wire-brushing using stainless wire-brush. The stacked sheets were spot-welded at the front and rear ends, then heated in an electric furnace set at 400°C for 600 s, and roll-bonded by 35% reduction in thickness in one pass. A two-high mill having the roll diameter of 310 mm was operated at a roll peripheral speed of 0.17 ms^{-1} without lubrication. The roll-bonded sheet was again held at 400°C for 600 s, and rolled by 24% reduction, so that the total reduction in thickness became 50% (von Mises equivalent strain of 0.8). The 50% rolled sheet was cut into two and the ARB procedure described above was repeated up to 5 cycles corresponding to total equivalent strain of 4.0. The materials ARB processed by 5 cycles were filled with the elongated submicrometer grains as will be shown later. In order to change the grain size, the sheets ARB processed by 5 cycles were annealed at various temperatures ranging from 500°C to 700°C for 1.8 ks in argon atmosphere. In order to make the final heat treatment condition for P segregation even, additional heat treatment at 400°C, at which P-segregation is enhanced to occur, for 6 ks was conducted for all the specimens including the starting sheets and the as-ARB processed sheets of both IF steels.

The microstructures of the specimens were observed on longitudinal sections of the sheets by optical microscopy, scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The thin foils perpendicular to the transverse direction (TD) of the sheets for TEM observations were prepared by twin-jet electropolishing in a 100 mL HClO_4 + 900 mL CH_3COOH solution at 11°C. Hitachi H-800 was operated at 200 kV for TEM. The mean grain size was determined as the mean intercept length on the micrographs. Because the as-ARB processed specimens and those annealed at low temperatures revealed the UFGs elongated along the rolling direction (RD), the grain thickness (d_t) parallel to the normal direction (ND) of the sheet and the grain length (d_l) parallel to RD were determined for such samples. Tensile tests of the specimens were carried out using 1/5 miniaturized specimen of JIS-5, which has gage width and length of 5 mm and 10 mm, respectively, at an

Fig. 1 The sub-size specimen for the miniaturized Charpy impact test.

initial strain rate of $8.3 \times 10^{-4} \text{ s}^{-1}$. The tensile direction was parallel to RD. The present ARB process produced the sheet samples 1 mm thick, so that it was impossible to do the standard Charpy impact test. Therefore, sub-sized specimens were cut from the sheets as is illustrated in Fig. 1 and tested at various temperatures from liquid nitrogen temperature to ambient temperature in the miniaturized Charpy impact machine developed by Kimura *et al.*¹²⁾ The absorbed energy in dynamic fracture was calculated from the displacement-load data recorded during the test. Fracture surface of the specimens was observed in a JEOL JSM5600 SEM operated at 20 kV.

3. Results

3.1 Microstructures of the materials

Figures 2(a) and (b) show the TEM microstructures of the IF steels ARB processed up to strain of 4.0. Both specimens reveal the lamellar boundary structure,¹⁷⁾ which has been observed in the IF steels ARB processed at 500°C.^{9,18–22)} The spacing of the lamellar boundaries in the P-added IF steel ($d_l = 0.18 \mu\text{m}$) was smaller than that in the plain IF steel ($d_l = 0.24 \mu\text{m}$). This corresponds with the recently reported result that the microstructure of the Al-2%Cu alloy first solution-treated and ARB processed was much finer than that of the 99% commercial purity aluminum.²³⁾ That is, solute atoms refine the deformation induced ultrafine microstructures not only in aluminum alloys but also in ferritic steels. The selected area diffraction (SAD) patterns taken by the use of an aperture 9 μm in diameter were not spotty but ring-like in both materials, indicating that the ultrafine lamellar boundary structures involve large local misorientations. Actually, it has been quantitatively clarified by electron backscattering diffraction (EBSD) analysis in SEM equipped with field-emission (FE) gun that the fraction of high-angle grain boundaries is above 75% in the plain IF steel ARB processed up to strain of 5.6 at 500°C.²²⁾ The quantitative analysis by FE-SEM/EBSD has also clarified that the microstructure is quite uniform throughout thickness of the ARB processed sheet.²²⁾ Although the present total strain (4.0) was smaller than the case mentioned above, it has been also shown in an IF steel that most of the lamellar boundaries are already high-angle after 5 cycles (strain of 4.0) of the

Fig. 2 TEM microstructures and corresponding SAD patterns of the plain IF steel (a,c) and the P-added IF steel (b,d). Observed from TD of the sheets. (a,b) As-ARB processed up to 4.0 of strain at 400°C. (c,d) ARB processed at 400°C and subsequently heat treated at 400°C for 6 ks to equalize the grain boundary segregation condition.

ARB at 500°C.²⁰⁾ It is reasonable, therefore, that the uniform ultrafine microstructures with large misorientations formed in the present materials by the ARB. The present microstructures, however, include high density of dislocations and they are not pancake-shaped but rather elongated straight, compared with the UFG structures previously reported.^{9,18–22)} This is probably because the present ARB process was carried out at lower temperature (400°C) than that in the previous cases (500°C^{9,18–22)}). The microstructures after the heat treatment at 400°C for 6 ks to equalize the grain boundary segregation condition are shown in Figs. 2(c) and (d). Recovery during holding at 400°C decreased the dislocations inside the grains and grain boundary migration in short ranges occurred. As a result, the microstructures became much more grain-like, though they still have elongated morphologies. The mean grain thickness (d_t) increased to 0.40 μm and 0.24 μm for the plain IF steel and the P-added IF steel, respectively.

During subsequent annealing at various temperatures, the recovery and the grain growth proceeded furthermore. The TEM microstructures of both materials after annealing at various temperatures for 1.8 ks are shown in Fig. 3. Annealing above 575°C for the plain IF or above 600°C for the P-added IF promoted the nearly equiaxed grain structures free from dislocations inside. It is difficult to distinguish these microstructures (Figs. 3(b,c,f)) from typically recrystallized microstructures. The microstructural changes observed in the present study coincide with those previously reported in the aluminum^{7–9)} and IF steel^{9,20,21)} ARB processed and an-

nealed. The mean grain thickness and length are plotted as a function of the annealing temperature in Fig. 4. It can be also understood from Fig. 4 that P-addition inhibits the grain growth in annealing. Anyhow, the bulky samples having various mean grain sizes (grain thickness) ranging from 0.24 μm to 12 μm were successfully obtained through the ARB process followed by annealing.

3.2 Strength and ductility obtained from static tests

Figure 5 shows the stress-strain curves obtained from the static tensile test of the IF steels having various mean grain sizes. The mechanical properties are again quite similar to those previously reported in the plain IF steel ARB processed at 500°C and subsequently annealed.^{9,20)} The strength of the P-added IF steel was higher than that of the plain IF steel treated under the same conditions, probably because of solution hardening by P and the finer microstructure shown in Fig. 2. The as-ARB processed specimens²⁴⁾ showed very high strength 2~3 times higher than those of the starting specimens having conventionally coarse grained structures. The as-ARB processed specimens reached the maximum strength at very early stage of the tensile test, followed by macroscopic necking. Consequently, the uniform elongation was below a few percents and the total elongation was within 10%. The strength decreased with increasing the annealing temperature (*i.e.*, with increasing the grain size), but uniform elongation recovered only after the grain size increased over 1 μm. The limited uniform elongation in the ultrafine grained steels having ferrite single-phase structure has been ex-

Fig. 3 TEM microstructures of the plain IF steel (a,b,c) and the P-added IF steel (d,e,f) ARB processed up to 4.0 of strain at 400°C, then heat treated at 400°C for 6 ks, and annealed at various temperatures for 1.8 ks. Observed from TD. (a) Annealed at 550°C. (b) 575°C. (c) 600°C (d) 550°C (e) 600°C (f) 650°C.

plained in terms of the plastic instability.⁹⁾

3.3 Results from dynamic fracture tests

Figure 6 shows the absorbed energies of the starting materials having conventionally coarse-grained structures as a function of the test temperatures. The fracture surfaces observed by SEM are also indicated for several test conditions. It is well known that ferritic steels having BCC structure perform discontinuous transition from ductile fracture to brittle fracture at a certain low temperature,²⁵⁾ which has been understood on the basis of significant temperature dependence of Peierls potential in BCC crystals. Such a ductile-brittle transition was clearly recognized in the present miniaturized Charpy testing for the coarse grained materials. The fracture surface coincides well with the change in the absorbed energy: ductile fracture surfaces

characterized by dimples were observed in the specimens which were tested at higher temperatures and showed larger absorbed energies, while brittle fracture surfaces mainly exhibiting intergranular fracture appeared at lower temperatures showing very small absorbed energies. The ductile-brittle transition temperatures (DBTT) were -154°C and -115°C for the plain IF steel and the P-added IF steel, respectively. The higher DBTT in the P-added specimen suggests the enhancement of grain boundary embrittlement by grain boundary segregation of P. Actually, cleavage fracture, which was partly recognized in the plain IF steel, was hardly seen in the P-added IF, suggesting the grain boundaries were weakened by the addition of P.

The absorbed energy-test temperature curves of the IF steels having various mean grain sizes are shown in Fig. 7. Different from Fig. 6, the specimens did not show the sharp

Fig. 4 The mean grain sizes (grain thickness (d_t) and grain length (d_l)) of the IF steels ARB processed and annealed at various temperatures.

decrease in the absorbed energy except for the plain IF specimens with the coarse grain sizes of 8 μm and 10 μm . Basically, the absorbed energy continuously decreased with decreasing the test temperature. The absorbed energies at room temperature and those at liquid nitrogen temperature (-190°C) are plotted as a function of the grain size in Fig. 8. Concerning the toughness at room temperature (so-called upper shelf energy), the absorbed energy obviously tends to decrease with decreasing the grain size, though the data are fairly scattering. For the absorbed energy at -190°C , scattering is furthermore severe, and it is difficult to find clear tendencies. However, it seems that the absorbed energy once increases with decreasing the grain size and then decreases in the submicrometer grain size region below 1 μm . It might be noteworthy, anyway, that the absorbed energies at -190°C of the fine grained specimens are evidently larger than that of the conventionally coarse grained specimens having mean grain sizes over 10 μm .

Macroscopic appearance of the dynamic fracture surfaces are summarized in Fig. 9. It is obvious that so-called delamination or separation^{26–28)} occurred in many samples. Roughly speaking, the number of the delamination increased with decreasing the test temperature and with decreasing the grain size. In the specimens having the grain sizes above several microns, however, the number of delamination was rather limited at low temperature. On the other hand, the ultrafine grained specimens exhibited many delamination at low temperature. Similar frequent delamination has been reported in the UFG SM490-steel by Ohmori *et al.*¹¹⁾ As will be discussed later, these characteristic features in the macroscopic fracture surface should be correlated with the change in the absorbed energy shown in Fig. 7.

The SEM photographs showing microscopic features of the fracture surfaces of the plain IF steel and the P-added IF steel are shown in Fig. 10 and Fig. 11, respectively. Although the energy-temperature curves did not show discontinuous transition (Fig. 7), the microscopic fracture surface exhibited significant changes. At higher temperatures, ductile fracture

Fig. 5 Nominal stress-strain curves of the plain IF steel (a) and the P-added IF steel (b) ARB processed and annealed at various temperatures to change the grain size. The annealing temperatures and the mean grain sizes (grain thickness) are indicated in the figures.

surfaces characterized by dimples or elongated dimples were observed, while the specimens tested at very low temperatures indicated brittle fracture surfaces mainly composed of intergranular fracture. The transition from the ductile fracture surface to the brittle fracture surface was discontinuous at a certain temperature. In case of the plain IF specimen with grain size of 5.5 μm , for instance, showed ductile fracture surface at -180°C but grain boundary fracture at -190°C . The temperature at which the transition in the fracture surface occurred decreased with decreasing the grain size. And finally no brittle fracture was observed even at -190°C in the both materials having submicrometer grain sizes (Fig. 10(f) and Fig. 11(f)). The results of the microscopic fracture surface observations are summarized in Fig. 12. Ductile-brittle transition could be determined from the observations of the microscopic fracture surface, as mentioned above. The DBTT clearly decreases by grain refinement, and the area showing brittle fracture (hatched area) is larger in the P-added IF steel than that in the plain IF steel. The DBTT determined from the microscopic fracture surface was plotted as a function of minus square root of the grain size in Fig. 13. Linear relationship like Hall-Petch relation is recognized in the both materials. Two lines for two kinds of the IF steels are nearly parallel. It can be concluded that the ultra-grain refinement is significantly effective to decrease the DBTT in ferritic steels. Even at liquid nitrogen temperature, no brittle fracture occurs if the mean grain size is smaller than 5 μm or 2 μm in the present plain IF or the P-added IF, respectively.

Fig. 6 Absorbed energy of the starting specimens of the IF steels measured from the miniaturized Charpy impact test at various temperatures. The mean grain sizes were $15\mu\text{m}$ and $12\mu\text{m}$ for the plain IF steel and the P-added IF steel, respectively. The SEM microstructures showing typical fracture surfaces are also shown for several testing conditions.

Fig. 7 Absorbed energy of the plain IF steel (a) and the P-added IF steel (b) having various fine grain sizes. Measured from the miniaturized Charpy impact test at various temperatures.

Fig. 8 Absorbed energies at room temperature (a) and at -190°C (b) of the IF steels having various ultrafine grain sizes.

4. Discussions

The dynamic fracture toughness of the plain IF and P-added IF steels having UFG structures was investigated in this study systematically as a function of the grain size

ranging from 0.24 to $11\mu\text{m}$. The various grain-sized specimens were successfully prepared through the ARB and annealing process. One of the characteristic results of the present study was remarkable delamination on the macro-

Fig. 9 SEM photographs showing the macroscopic fracture surfaces of the IF steels after the miniaturized Charpy impact test.

scopic fracture surfaces, as was shown in Fig. 9. The 5 cycle ARB-processed sheet includes $2^5 = 32$ pieces of the initial sheet within the final thickness of 1 mm. The delamination, therefore, should be primarily considered with regard to the

31 bonded boundaries included within the present ARB processed sheets. In the relatively coarse-grained specimens fractured at relatively high temperatures, the number of the delamination was limited and the most severe separation often occurred at the thickness center of the sheet (Fig. 9). At the thickness center of the ARB processed sheet, there is the bonded boundary formed in the last cycle of the ARB, which is the weakest bonded boundary in the sample. In fact, separation at the last bonded boundary has been sometimes observed in the ARB sample after fracture in static tensile test. The next severe separation tended to occur at quarter thickness of the sheet, and the large delamination seemed to appear in regular intervals, which seems to correspond with the bonded boundaries regularly existing within the ARB processed sheets. That is, the frequent delamination in the impact tested samples may be a specific result in the ARB processed materials.

However, the regular appearance of delamination is typical even in bulk materials without any bonding.^{27–29)} It is noteworthy, moreover, that the number of delamination rather decreased at very low temperature like -190°C in the coarse grained specimens having the grain sizes larger than $2\mu\text{m}$ (Fig. 9). This agrees well with the result previously reported in bulk samples.²⁸⁾ After a detailed examination, it was actually found that the positions of the delaminations in the present ARB specimens did not necessarily coincide with those of the bonded boundaries. Furthermore, there are many small separations branched off from big ones, as is shown in Fig. 9. Ohmori *et al.*¹¹⁾ have recently reported a number of delaminations, which are quite similar to the present ones, in the ultrafine grained SM490 steel. Their UFG steel was produced by mutliaxial bar rolling at warm temperatures, so that there were no bonded boundaries within the material. Consequently, we conclude that the present delamination is not a specific feature of the ARB processed materials but it is

Fig. 10 Microscopic fracture surfaces of the plain IF steel. (a,b) The specimen with mean grain size of $11\mu\text{m}$. (c,d) Grain size of $5.5\mu\text{m}$. (e,f) Grain size of $0.44\mu\text{m}$. Testing temperatures: (a) -140°C . (b) -150°C . (c) -180°C . (d) -190°C . (e) -180°C . (f) -190°C .

Fig. 11 Microscopic fracture surfaces of the P-added IF steel. (a,b) The specimen with mean grain size of 10 μm . (c,d) Grain size of 2.2 μm . (e,f) Grain size of 0.76 μm . Testing temperatures: (a) -110°C (b) -120°C (c) -170°C (d) -180°C (e) -180°C (f) -190°C

Fig. 12 Relationship between the mean grain size and the microscopic fracture surfaces in the plain IF steel (a) and the P-added IF steel (b).

rather a characteristic feature of the UFG ferritic steels fabricated through heavy rolling deformation.

The occurrence of the delamination has been explained in terms of fiber textures. Matsuda *et al.*,²⁸⁾ reported that the

Fig. 13 The DBTT determined from the microscopic fracture surface of the IF steels as a function of minus square root of the mean grain size.

separation forms by the initiation of the crack along the interface between the colonies having $\{100\}$ texture and that having $\{111\}$ texture. The crack initiation is associated with the difference in plastic anisotropy between the $\{100\}$ and $\{111\}$ oriented crystal. The $\{100\}$ and $\{111\}$ orientations are the main components of the α -fiber and γ -fiber textures, respectively, which are the typical rolling textures in ferritic steels.³⁰⁾ Ohmori *et al.*¹¹⁾ also concluded that the delamination in their UFG steel is owing to the fiber texture developed during heavy warm rolling. In the present ARB processed specimens, there must be such textures. Though complicatedly distributed shear texture must be taken into account in case of the specimen ARB processed under high friction conditions,³¹⁾ Tsuji *et al.*¹⁹⁾ and Kamikawa *et al.*²²⁾ clarified by FE-SEM/EBSP analysis that the ARB processed IF steels involve the α -fiber and γ -fiber textures. Since the lamellar boundaries in the ARB processed materials have large

misorientations,^{18,19,22,32)} the regions having {100} texture, {111} texture and other orientations finely and mutually distributed throughout thickness of the sheet, which might be the reason for a number of fine delamination observed in the present specimens (Fig. 9). The textures in the as-ARB processed specimens mostly remain during annealing, because continuous recrystallization basically occurs in such materials.^{7,8,33)} In case of the starting materials which did not show the delamination, the microstructure was fully recrystallized grains with coarse grain sizes. It is reasonable to expect the {100} and {111} fiber textures are weak in the recrystallized material. Furthermore, intergranular fracture happened in such coarse-grained materials at low temperatures before the delamination initiates (Fig. 9).

Independent of the formation mechanism, the delamination must be associated with the absorbed energy in the impact test. The continuous decrease in the absorbed energy with decreasing the test temperature in the ARB processed and annealed specimens (Fig. 7) is attributed to the increase in the number of delamination. The upper shelf energy at ambient temperature (Fig. 8(a)) decreased with decreasing the grain size. This also corresponds to the increase in number of delamination on the macroscopic fracture surface at room temperature, as is shown in Fig. 9. It can be concluded, therefore, that ultra-grain refinement reduces the upper shelf energy of ferritic steels in dynamic fracture, because of the occurrence of delamination. Since heavy deformation is necessary to realize the UFG structures in both ultimate TMCP and SPD processes, this decrease in upper shelf energy would not be avoidable in the UFG ferritic steels. However, even in the P-added IF steel with 0.2 μm grain size the absorbed energy was larger than 50% of the coarse grained specimens.

It is noteworthy, furthermore, that the absorbed energies of the UFG specimens at -190°C were higher than those of the coarse grained specimens, as was shown in Fig. 8(b). Especially down to the grain size around 0.5 μm , the absorbed energy at very low temperature tended to increase with decreasing the grain size, although the number of delamination increased (Fig. 9). It should be noted that the specimens with the grain sizes smaller than 0.5 μm showed elongated grain morphologies and involved high density of dislocations inside (Fig. 3). That is, the decreasing absorbed energy in the specimens whose grain sizes are smaller than 0.5 μm might be due to the dislocation substructures included in the materials. Ultra-grain refinement of ferrite is likely to improve the toughness at low temperatures essentially.

The microscopic fractographies obviously changed by grain refinement. The temperature range at which brittle fracture (mainly grain boundary fracture) surface appeared shrunk with decreasing the grain size, as was shown in Fig. 12. The DBTT determined from the microscopic fracture surface had a Hall-Petch like relationship with the grain size (Fig. 13). It is noteworthy that no brittle fracture was observed even at liquid nitrogen temperature,³⁴⁾ if the mean grain size was smaller than 5 μm in the plain IF steel or smaller than 2 μm even in the P-added IF steel. It has been reported that the delamination grow in cleavage mode,²⁶⁾ and this accommodates the three-axial stress, resulting in the decrease in the transition temperature.^{11,29)} Together with this

delamination effect, however, the effect of the structural refinement should be taken into account for the toughness improvement as well. The ultra-grain refinement greatly increases the strength of the material itself, as was shown in Fig. 5. The UFG structure would decrease the stress concentration at grain boundaries, especially at triple junctions, because the number of them significantly increases in the UFGs.²⁾ It can be concluded, therefore, that ultra-grain refinement of ferrite inhibits the intergranular fracture to improve the dynamic fracture toughness at low temperature.

The DBTT of the starting plain IF steel having grain size of 15 μm was -154°C (Fig. 6). As was described before, the DBTT greatly decreased with decreasing the grain size. According to Fig. 13, the DBTT is below -150°C even in the P-added IF steel when the grain size is smaller than 5 μm . That is, as far as the DBTT is concerned, phosphorus seems to be made harmless by grain refinement. In the present study, the direct measurement of the P concentration at grain boundaries was not carried out. However, we consider that the concentration of P was not decreased so much in the discussing grain size range above 2–5 μm , since the decrease in the DBTT of the P-added IF steel with decreasing the grain size was nearly parallel to that of the plain IF steel (Fig. 13). This agrees with the argument by Takaki³⁶⁾ that the grain boundary segregation of P is not so much suppressed if the grain size is larger than 5 μm . That is, the decrease in the DBTT of the P-added steel was attributed to the effect of grain refinement itself. On the other hand, the amount of P segregated at grain boundaries might be greatly suppressed even in the present specimens having submicrometer grain sizes.

5. Conclusions

Dynamic fracture toughness of the ultrafine grained plain IF steel and P-added IF steel was studied systematically as a function of the grain size in the range from 0.24 to 11 μm . The major results obtained are as follows:

- (1) The ultrafine grained structures whose mean grain sizes range from 0.24 to 11 μm could be successfully obtained through the ARB process followed by annealing. It was clarified that solute P is effective to refine the deformation induced ultrafine structures. The grain growth of the UFGs during annealing was also inhibited by P-addition.
- (2) The static strength of the materials significantly increased with decreasing the grain size and it reached to 750 MPa and 820 MPa in the IF steel and the P-added IF steel, respectively, having submicrometer grain sizes. On the other hand, the uniform elongation of the ultrafine grained steels was limited within a few percents probably due to early plastic instability.
- (3) A number of delamination happened at low temperatures in the impact-tested specimens especially having ultrafine grained structures. The delamination was not necessarily owing to roll-bonding in the ARB process but it is rather a characteristic feature of the ultrafine grained ferritic steels.
- (4) The upper shelf energy in the impact test at ambient temperature tended to decrease with decreasing the

grain size. On the other hand, the ultrafine grained specimens showed higher absorbed energies than the coarse grained specimens at liquid nitrogen temperature.

- (5) Microscopic fracture surface exhibited a sudden transition from ductile fracture characterized by dimples into brittle fracture mainly due to intergranular fracture at a certain low temperature. The ductile-brittle transition temperature determined from the microscopic fractography greatly decreased with decreasing the grain size, and finally no brittle fracture occurred even at -190°C when the mean grain size was smaller than $5\mu\text{m}$ or $2\mu\text{m}$ in the plain IF steel or the P-added IF steel, respectively. Consequently, ultra-grain refinement is effective to improve toughness of ferritic steels, and phosphorus can be made substantially harmless by ultra-grain refinement.

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