1. Introduction

In the past decade, extensive researches have been devoted to fabrication and characterization of ultrafine grained (UFG) materials with the advent of severe plastic deformation (SPD) techniques. In spite of ultrahigh strength of UFG materials, a lack of strain hardenability and corresponding low uniform tensile elongation are the primary mechanical drawbacks of UFG materials, and therefore their practical application to the structural use is very limited at present. Accordingly, recent investigations in this area are focused on ductilization and restoration of strain hardenability of UFG materials, which other UFG materials hardly exhibit, in addition to ultrahigh strength and good uniform elongation. By selecting the optimum ECAP and intercritical annealing conditions, it was possible to fabricate UFG F/M DP steel in which isolated UFG martensite islands were uniformly embedded into UFG ferrite matrix. The formation of such a unique microstructure under the present processing conditions was discussed in terms of microstructural evolution during ECAP. Room temperature tensile properties of UFG F/M DP steel were superior to those of coarse grained counterpart. More importantly, in spite of an UFG structure, the present UFG F/M DP steel exhibited extensive strain hardenability from the onset of plastic deformation in association with grain size independent strain gradient plasticity, unlike other UFG materials.

KEY WORDS: ultrafine grain; dual phase steel; equal channel angular pressing; microstructure; tensile properties.

2. Fabrication of UFG F/M DP Steel

The present UFG F/M DP steel was processed by equal channel angular pressing (ECAP) and subsequent intercritical annealing was described in detail. Along with strain gradient plasticity concept in which an introduction of a high density of geometrically necessary dislocations determines to enhance strain hardenability, the aim of the present study was fabrication of UFG F/M DP steel exhibiting extensive strain hardenability, which other UFG materials hardly exhibit, in addition to ultrahigh strength and good uniform elongation. By selecting the optimum processing variables of each processing step, it was possible to fabricate UFG F/M DP steel in which isolated UFG martensite islands were uniformly embedded into UFG ferrite matrix. The formation of such a unique microstructure under the present processing conditions was discussed in terms of microstructural evolution during ECAP. Room temperature tensile properties of UFG F/M DP steel were superior to those of coarse grained counterpart. More importantly, in spite of an UFG structure, the present UFG F/M DP steel exhibited extensive strain hardenability from the onset of plastic deformation in association with grain size independent strain gradient plasticity, unlike other UFG materials.

KEY WORDS: ultrafine grain; dual phase steel; equal channel angular pressing; microstructure; tensile properties.
cessing step in order to maintain UFG structure since intercritical annealing is undertaken at high temperatures belonging to ferrite/austenite two phases region. So, in this section, each processing step is described in detail.

2.1. Material

A plain carbon–manganese steel (Fe–0.15C–0.25Si–1.1Mn (in mass%)) was used in the present investigation. It was prepared as a 50 kg ingot by a vacuum induction melting. The ingot was homogenized at 1250°C for 1 h and size-rolled to the plate of 50 mm thickness and 150 mm width. The rolled plate was austenitized at 1200°C for 1 h and then air-cooled.

2.2. Equal Channel Angular Pressing

ECAP was performed on the cylindrical samples of 130 mm length and 10 mm diameter which were machined from the rolled plate. The outer curvature and inner contact angles of intersection between two channels of the present ECA pressing die were 20° and 90° respectively so that it yielded an effective strain of ~1 by a single pass. Each sample was subjected to 4 passes, i.e. total accumulated effective strain of ~4, since structural refinement became saturated at this level of ECAP strain. During ECAP, the sample was rotated 180° around its longitudinal axis between the passages, i.e. route C. Route C was selected by the following considerations: (a) it restores the shape of the original segment at even number passes and thereby a nearly equiaxed UFG structure can be obtained, and (b) it produces higher effective strain without changing the shape at even number passes than other ECAP routes due to repetitive shearing on the same plane. ECAP was conducted at 500°C with a ram speed of 2 mm/s. The relatively high ECAP temperature was selected in order to minimize catastrophic abnormal grain growth of UFG retained ferrite during subsequent intercritical annealing and to accelerate the diffusion kinetics of carbon atoms dissolved from pearlitic cementite into UFG ferrite matrix. As discussed later, the latter was the crucial phenomenon to homogeneous distribution of UFG martensite islands through UFG ferrite matrix.

2.3. Intercritical Annealing

Of the conventional methods fabricating F/M DP steel, a heat treatment in which ferrite/pearlite steel is heated up in the range of ferrite/austenite two phases region, i.e. between $A_{c1}$ and $A_{c3}$ temperatures, is referred as intercritical annealing. Under a fixed chemical composition, time and temperature of intercritical annealing are the critical factors determining the microstructural, compositional and mechanical characteristics of constituent phases and so whole mechanical properties of F/M DP steel.

2.3.1. Determination of Intercritical Annealing Temperature

The microstructure of UFG metallic materials fabricated by SPD including ECAP is thermally unstable due to a large accumulation of strain energy. This causes catastrophic abnormal grain growth of ultrafine grains—for the present case, UFG retained ferrite grains—during heat treatment. Therefore, intercritical annealing temperature is desired to be as low as possible, i.e. just above $A_{c1}$ temperature, to avoid abnormal grain growth. Accordingly, $A_{c1}$ temperature of the present ECAPed UFG ferrite/pearlite steel was determined precisely by a dilatometry experiment with a heating rate of 10°C/s. A comparison of dilatation curves between ECAPed UFG ferrite/pearlite steel and unECAPed coarse grained ferrite/pearlite steel was made in Fig. 1. It was found that ECAP did not affect either $A_{c1}$ (725°C) or $A_{c3}$ (870°C) temperatures. However, the former showed a rapid negative dilatation at the onset of reverse austenite transformation, indicating that ECAP altered transformation kinetics in association with local compositional change due to carbon atom dissolution from pearlitic cementite and high accumulated strain energy during ECAP. Based on the dilatometry experiment, the present intercritical annealing was carried out at 730°C which was 5°C higher than $A_{c1}$ temperature of the ECAPed UFG ferrite/pearlite steel. The room temperature water (cooling rate >300°C/s) was used as a quenching medium.

2.3.2. Determination of Intercritical Annealing Time

The two considerations were made to determine intercritical annealing time. First, the resultant martensite volume fraction after quenching corresponds to the equilibrium austenite volume fraction of the present steel at intercritical annealing temperature of 730°C to ensure the completion of reverse austenite transformation. A thermodynamic analysis revealed that the equilibrium austenite volume fraction of the present steel at intercritical annealing temperature of 730°C was ~27.4%. Second, with satisfying the first requirement, intercritical annealing time is desired to be as short as possible to avoid abnormal grain growth as mentioned above. A series of preliminary tests revealed that over 10 min annealing at 730°C resulted in abnormal grain growth in some areas of the sample. Therefore, the intercritical annealing time was fixed as 10 min through the experiment.

![Fig. 1. A comparison of dilatation curves (a heating rate of 10°C/s) between ECAPed UFG ferrite/pearlite steel and unECAPed coarse grained ferrite/pearlite steel.](image-url)
3. Microstructures

3.1. Before Intercritical Annealing

Microstructures of the present steel before and after ECAP are compared in Fig. 2. The macrostructure before ECAP (Fig. 2(a)) consisted of approximately 20 vol% pearlite (dark patches) and the remainder of ferrite. Both ferrite grains and pearlite colonies were nearly equiaxed and their size was about 30 μm. Dislocations with moderate density existed in ferrite (Fig. 2(c)), and pearlite consisted of a well developed lamellar structure (Fig. 2(e)). Meanwhile, after ECAP, pearlite colonies were severely deformed and fragmented into smaller ones with a very irregular shape (Fig. 2(b)). Macroscopically, this resulted in decrease of inter-pearlite colony spacing. Ferrite grains were significantly refined to 0.2–0.5 μm with a dense dislocation structure (Fig. 2(d)). Cementite lamellar plates were severely deformed and became thinner with ECAP, and even some extent of spheroidization of cementite occurred (Fig. 2(f)).

3.2. After Intercritical Annealing

3.2.1. Microstructural Characteristics of UFG F/M DP Steel

The microstructure of UFG F/M DP steel, which resulted from intercritical annealing and quenching of UFG ferrite/pearlite steel, consisted of ferrite grains and uniformly distributed martensite islands (Fig. 3(a)). Each martensite island was in an isolated blocky type (Fig. 3(b)). A careful measurement using scanning electron microscopy (SEM) and an image analyzer revealed that both ferrite grain size and martensite island size were ~0.8 μm and the martensite volume fraction was about 28%. As mentioned above, the present martensite volume fraction was very close to the equilibrium austenite volume fraction at intercritical annealing temperature of 730°C, demonstrating that 10 min intercritical annealing was just enough to the completion of reverse austenite transformation. A very high density of dislocations was observed in the ferrite grain adjacent to martensite (Fig. 3(c)). These dislocations were generated in order to accommodate transformation-induced-strain built between martensite transformed by quenching and retained ferrite. In addition, they are known to be mobile and play an important role on rapid, extensive strain hardening of F/M DP steel from the onset of its plastic deformation.5,9) It is worth mentioning that dislocations generated by ECAP are expected to be rapidly annihilated due to relatively high ECAP and intercritical annealing temperatures. Figure 4 shows the distribution of grain boundary misorientation of ferrite in UFG F/M DP steel. A fraction of low angle boundaries, i.e. less than 15°, was about 58%.

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![Fig. 2. Microstructures of unECAPed coarse grained ferrite/pearlite steel (a, c, and e) and ECAPed UFG ferrite/pearlite steel (b, d, and e): (a), (b) optical micrographs, (c), (d) TEM micrographs of ferrite, (e), (f) TEM micrographs of pearlite.](image-url)
3.2.2. Formation of UFG F/M DP Steel

In general, conventional coarse grained F/M DP steels prepared by intercritical annealing exhibit the large irregular volumes of martensite transformed from austenite which are formed by pearlite decomposition during intercritical annealing, so mostly residing at the former pearlite colonies,⁹ as shown in Fig. 5: the identical chemical composition and processing conditions, but without ECAP, were used for fabrication of coarse grained F/M DP steel in Fig. 5. A comparison of Figs. 3(a) and 5 revealed significant differences of the characteristics of martensite, i.e. distribution, size, shape and sites, between UFG F/M DP and coarse grained F/M DP steels.

In order to achieve an unique microstructure of the present UFG F/M DP steel, i.e. uniform distribution of isolated UFG martensite islands throughout UFG ferrite matrix, at least the two necessary conditions should be satisfied: (a) uniform distribution of potential austenite nucleation sites, and (b) the carbon content of those potential austenite nucleation sites equivalent to the equilibrium carbon content in austenite at intercritical annealing temperature. The first requirement could be fulfilled with microstructural evolution during ECAP. The microstructure of the ECAPed ferrite/pearlite steel is manifested by subdivided pearlite colonies, UFG ferrite grains with a high density of lattice dislocations, and a large number of low- and high-angle boundaries with extrinsic boundary dislocations, etc.¹¹ All these factors are anticipated to provide uniformly distributed austenite nucleation sites.

The second requirement is associated with dissolution of carbon atoms from pearlitic cementite and their concurrent diffusion to potential austenite nucleation sites during ECAP.¹²–¹⁴ Although no quantitative data for cementite dissolution during ECAP is at present available, an indirect information can be obtained from the severely cold drawn pearlitic steel wire¹⁵–¹⁸ since (a) the reported maximum drawing strain ~5, was comparable to the present ECAP strain, ~4, and (b) the morphological evolution of pearlitic cementite during ECAP was similar to that observed in the severely cold drawn pearlitic steel wire. The previous investigations¹⁷,¹⁸ reported that 20–50% of pearlitic cementite was dissolved during cold drawing and resultantly the carbon content in ferrite reached over ~4 at%.

From Fig. 2(f) showing dissolved pearlitic cementite lamellar and the present ECAP strain, a similar carbon content is expected to exist in pearlitic ferrite and in UFG ferrite in the vicinity of pearlite colonies after ECAP. Then, as a first approximation, the distribution of the carbon content in ferrite matrix

Fig. 3. Microstructures of UFG F/M DP steel. (a) SEM micrograph showing distribution of martensite islands. (b) Enlarged SEM micrograph showing the shape and size of martensite islands. (c) TEM micrograph showing the presence of dislocations with high density in the vicinity of martensite islands.

Fig. 4. A histogram showing the distribution of grain boundary misorientation angle of ferrite in UFG F/M DP steel.

Fig. 5. Microstructure of coarse grained F/M DP steel fabricated by intercritical annealing without ECAP (white patches are martensite): chemical composition and intercritical annealing conditions were identical with those of UFG F/M DP steel.
can be estimated by applying the diffusion solution for a pair of semi-infinite solids,\textsuperscript{19) i.e.}
\[ C(x,t) - C_o = 1 - \text{erf} \left( \frac{x}{\sqrt{4Dt}} \right) \]  
(1)

For the present case, \( C(x,t) \) is the carbon content in ferrite at the diffusion time \( t \) and diffusion distance \( x \) from pearlite colony, \( C_o \) is the carbon content in ferrite remote from pearlite colony, \( C_1 \) is the carbon content supersaturated in pearlitic ferrite, and \( D \) is the carbon diffusivity in ferrite at the ECAP temperature. The use of Eq. (1) may not lead to significant erroneous approximation since \( C_1 \) is not expected to vary significantly for short time diffusion: that is, carbon atoms are continuously supplied from remaining severely deformed pearlitic cementite. The following values were used for estimation: \( C_o = 0.09 \text{ at}\% \) (the equilibrium carbon content in ferrite at 500°C), \( C_1 = 4.0 \text{ at}\% \) (the reported carbon content supersaturated in pearlitic ferrite of severely cold drawn pearlitic steel\textsuperscript{18}), and \( D = 0.62 \times 10^{-6} \exp(-80,400 / (\text{J/mol}) / kT) \text{ m}^2/\text{s} \) (carbon diffusivity in \( \alpha \)-ferrite). The profile of the carbon content at various diffusion times is shown as a function of the distance from the pearlite colony in Fig. 6. The equilibrium carbon content in austenite at the intercritical annealing temperature calculated by a thermodynamic simulation, 2.38 at\%, is also presented in Fig. 6. The carbon content reaches the equilibrium carbon content in austenite at the intercritical annealing temperature at about 10, 23, and 33 \( \mu \text{m} \) away from pearlite colony for 1, 5 and 10 min, respectively. These distances are comparable to or larger than a half of the inter-pearlite colony spacing in the present UFG ferrite/pearlite steel (Fig. 2(b)). The total ECAP processing time was much longer than 10 min. The above estimation informs that reverse austenite transformation is likely to occur at the potential sites distributed uniformly throughout UFG ferrite matrix during intercritical annealing, and therefore uniformly distributed UFG martensite islands are formed by quenching.

4. Room Temperature Tensile Properties

The representative engineering stress–strain curves of as-ECAPed UFG ferrite/pearlite steel (Fig. 2(b)), UFG F/M DP steel (Fig. 3(a)) and coarse grained F/M DP steel (Fig. 5) are compared in Fig. 7 and their tensile data are listed in Table 1. Like other UFG metallic materials, UFG ferrite/pearlite steel exhibited no strain hardening, i.e. elastic-perfect plastic behavior, due to the ferrite grain size comparable to the dislocation mean free length.\textsuperscript{21) By contrast, in spite of UFG ferrite grains, UFG F/M DP steel exhibited continuous yielding, low yield ratio (YS/UTS), and rapid extensive strain hardening at the initial stage of plastic deformation, similar to coarse grained F/M DP steel. However, YS and UTS of UFG F/M DP steel were much higher than those of coarse grained F/M DP steel in spite of almost the same uniform elongation (\( \epsilon_u \)) and even larger total elongation (\( \epsilon_f \)). An excellent combination of ultrahigh strength, good ductility and extensive strain hardening of the present UFG F/M DP steel is attributed to the combined effects of (a) UFG ferrite grains responsible to ultrahigh strength and (b) existence of fresh mobile dislocations with high density ensuring extensive strain hardening and corresponding good uniform elongation. More detailed explanation for strain hardenability of UFG F/M DP steel is described elsewhere.\textsuperscript{22)} As demonstrated in Introduction, the present results suggest that transformation accommodation

Table 1. The nominal tensile properties of as-ECAPed UFG ferrite/pearlite steel, UFG F/M DP steel, and coarse grained F/M DP steel.

| designation            | YS , MPa | UTS , MPa | \( \epsilon_u \), % | \( \epsilon_f \), % | YS/UTS |
|------------------------|----------|-----------|---------------------|----------------------|---------|
| UFG ferrite/pearlite   | 605      | 648       | 9.8                 | 21.7                 | 0.93    |
| UFG F/M DP             | 540      | 890       | 9.8                 | 17.6                 | 0.61    |
| coarse grained F/M DP  | 504      | 764       | 10.3                | 13.5                 | 0.66    |
dislocations, which are generated by embedding hard martensite into soft ferrite matrix and regarded as geometrically necessary dislocations, are useful to store strain hardenability of UFG steels by inducing strain gradient plasticity which is independent of the ferrite grain size.

5. Conclusions

(1) Ultrafine grained ferrite/martensite dual phase (UFG F/M DP) steel was successfully fabricated by applying a combined process of severe plastic deformation via equal channel angular pressing (an effect strain of $\sim$4, route C and 500°C) and subsequent intercritical annealing (730°C×10 min).

(2) Microstructure of the present UFG F/M DP steel consisted of UFG ferrite matrix with homogeneously distributed UFG martensite islands. Ferrite grain size and martensite island size were $\sim$0.8 μm.

(3) Room temperature tensile properties of UFG F/M DP steel were superior to those of coarse grained counterpart. More importantly, in spite of an UFG structure, the present UFG F/M DP steel exhibited extensive strain hardenability from the onset of plastic deformation by the presence of mobile transformation accommodation dislocations with high density inducing grain size independent strain gradient plasticity.

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