Ultrafine Grained Low Carbon Steels Fabricated by Equal Channel Angular Pressing: Microstructures and Tensile Properties

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Equal channel angular pressing (ECAP) was conducted on the two grades of low carbon steel, with or without vanadium, in order to produce an ultrafine grained structure. As a result, the ferrite grains were refined from 30 μm to 0.2–0.3 μm. The strength of the ECAPed steels increased remarkably, over twice of the strength of the steels before ECAP. A series of static annealing experiments showed that the increment of ECAP strain and the dilute addition of microalloying element such as vanadium were very effective on enhancing thermal stability of the ultrafine gained low carbon steels produced by ECAP in terms of microstructure and tensile properties. This enhanced thermal stability resulted from; (a) presence of excessive carbon content in the ferrite matrix by carbon dissolution from pearlitic cementite during ECAP; (b) preservation of high dislocation density due to addition of vanadium, providing the effective diffusion path for dissolved carbon atoms; (c) precipitation of excessive carbon as the form of nano-sized cementite particles during subsequent annealing and its effect on suppressing grain growth.

KEY WORDS: low carbon steels; ultrafine grains; severe plastic deformation; equal channel angular pressing; microstructure; tensile properties.

1. Introduction

The extrapolation of the grain size dependency of the mechanical properties of metals and alloys to the extremely small size predicts an excellent combination of ultrahigh strength and toughness that cannot be achievable by other strengthening mechanisms. This leads to the intensive researches on fabrication of ultrafine grained (UFG) or nanostructured materials and their characterization in the last decade.1) Of several processing routes for fabricating UFG materials, severe plastic deformation (SPD), at present, is the most developed and viable method in terms of an ability producing fully-dense bulk UFG materials.2) Processing of fully-dense bulk UFG materials is essential not only to practical application in a large scale but also to consistent evaluation of their inherent properties. On the other hand, the recent increasing trend of effective construction of gigantic infrastructure, such as skyscrapers, long-span bridges, etc., demands stronger and tougher low carbon steels as the most widely used structural material. Accordingly, several investigations have been performed to fabricate UFG low carbon steels with ultrahigh strength by SPD.3–5) However, the microstructures of UFG materials produced by SPD, including low carbon steels, are known to be thermally unstable due to their non-equilibrium nature.6–8) This implies that grain growth easily occurs in these materials, causing the loss of mechanical superiority associated with an UFG structure. Therefore, for full utilization of UFG low carbon steels produced by SPD, it is essential to ensure a thermal stability. Along with the motivations addressed above, the present investigation was performed: (a) to clarify the grain refinement mechanism in the low carbon steels during SPD, (b) to examine the microstructural evolution of low carbon steel during SPD and subsequent heat treatment and the corresponding tensile properties, and (c) to find feasibilities to obtain a thermally stable UFG low carbon steels.

2. Experimental

2.1. Starting Materials

In the course of investigation, the two grades of low carbon steel were used: Fe–0.15C–0.25Si–1.1Mn (in wt%) (hereafter, CS steel) and Fe–0.15C–0.25Si–1.1Mn–0.06V–0.008N (in wt%) (hereafter, CSV steel), were used. Both steels were prepared as 50 kg ingots in the vacuum induction furnace. Ingots of both steels were homogenized at 1,523 K for 1 h and then size-rolled to fabricate the plates of 350×150×50 (mm²). The CS steel was austenitized at 1,523 K for 1 h and then air-cooled. The CSV steel was oil-quenched to room temperature after the same austenitization treatment as the CS steel and then normalized.
Normalization for the CSV steel consisted of soaking at 1 223 K for 1 h and the subsequent air-cooling. After heat treatment, a typical ferrite–pearlite structure was obtained in both steels. The ferrite grain size was ~30 μm and ~10 μm for the CS and CSV steels, respectively. Pearlite colony size was comparable to that of ferrite in both steels.

2.2. SPD Process: Equal Channel Angular Pressing

Equal channel angular pressing (ECAP) was used as a SPD process. In ECAP, a sample is pressed repeatedly through a die containing two channels, with equal cross-sections, intersecting at a certain angle so that the initial sample dimension is not altered by repetitive pressing.9,10 It is well established that ECAP can produce bulk UFG materials of the grain size down to ~150 nm. The present ECAP die was designed to yield an effective strain of ~1 per pass: the inner contact angle and the arc of curvature at the outer point of contact between channels of the die were 90° and 20°, respectively.11 During ECAP, the sample was rotated by 180° around its longitudinal axis between the passages. This pattern of pressing is commonly designated as route C.10,12 After machining the cylindrical samples of 18 mm φ×130 mm from the heat-treated plates of both steels, ECAP was carried out on the samples at 623 K up to 8 passes.

2.3. Post Heat Treatment and Tensile Test

In order to examine thermal stability of the ECAPed steels, 1 h static annealing was conducted at temperatures of 373–873 K using silicone oil bath or molten salt bath and the resultant microstructural changes were examined by field emission scanning electron microscope (FE-SEM, JEOL6330F) and transmission electron microscope (TEM, JEOL2010). Room temperature tensile tests were performed on the samples with the gage length of 25.4 mm using an Instron machine (Model 1125) at an initial strain rate of 1.33×10⁻³ s⁻¹.

3. Results and Discussion

3.1. Microstructural Evolution

3.1.1. As-ECAPed State

Figure 1 shows TEM micrographs of the CS steel by the ECAP passage. After 4-passes ECAP, the ferrite grains with
the initial size of $\sim 30 \mu m$ (Fig. 1(a)) were refined to 0.2–0.3 $\mu m$ (Fig. 1(b)). Further repeating ECAP up to 8 passes did not result in significant grain refinement (Fig. 1(c)), but an inspection of the corresponding selected area diffraction pattern revealed that it contributed to an increase in the misorientation angle across the grain boundaries. The morphological change in pearlite by the ECAP passage is shown in Figs. 1(d) to 1(f). Pearlitic cementite became more severely curled with increasing the ECAP passage. This capability of cementite for plastic deformation is very uncommon since cementite is brittle due to the lack of the number of operative slip system for homogeneous deformation, i.e. at least five, under the uniaxial stress condition.7) The similar morphological change of cementite during severe plastic working can be observed in the cold drawn eutectoid steel with full pearlite structure.13,14) In cold drawing of the eutectoid steel, plastic deformability of cementite is explained by operation of the additional slip systems under the hydrostatic stress state.15–17) The principal deformation mode in ECAP is thought to be either pure shear18) or shear with concurrent rotation. At this stage, it is unclear how cementite deforms plastically under shear mode and this issue might be further investigated.

In order to examine the ferrite grain reﬁnement mechanism, the detailed analysis of the slip systems operating in the ﬁrst two ECAP passages was conducted. As shown in Fig. 2(a) which was taken with the (111) zone axis, thin parallel shear bands were formed and their boundaries were aligned along the specific direction by the ﬁrst pass. There also exist many dislocation cell boundaries having different angles with band boundaries inside shear bands. By the second pass, the parallel band boundaries became serrated, as shown in Fig. 2(b), indicating operation of other slip systems which were not active in the ﬁrst pass. For the identiﬁcation of the slip systems, the angular relationship between band or cell boundaries was carefully analyzed and the result is also shown schematically in Fig. 2: the detailed method identifying the slip system is described in refs. 19, 20). As shown in Fig. 2, all slip systems operating in the ﬁrst two passes of ECAP with route C belong to the typical slip systems of BCC structure, i.e., \{111\}{110}, \{111\}{112} and \{111\}{123}. Very recently, Fukuda et al.4) reported the same observation on the sample processed with route Bc. Route Bc in which the sample is rotated by 90° around its longitudinal axis in the same direction between each passage is known to result in more homogeneous microstructure than other pressing routes of ECAP.21) The present analysis of the slip characteristics clearly reveals that the ferrite grain reﬁnement by ECAP is attributed to mechanical fragmentation associated with operation of multi slip systems.

3.1.2. Annealed State

Many previous investigations6,7,22) revealed enhanced grain growth behavior of ECAPed metals and alloys compared to those produced by conventional working processes due to relatively large accumulated strain. For full utilization of advantages arising from an ultraﬁne grain structure, it is essential to ensure thermal stability of these materials. For this purpose, the two approaches were attempted to improve thermal stability of the ECAPed steels: the increment of ECAP strain and microalloying. The first concept is related to the fact that carbon dissolves from pearlitic cementite by severe working such as cold drawing and excessive carbon higher than the equilibrium content exists in ferrite.23,24) It would cause retardation of recovery, recrystallization and grain growth. In addition, carbon dissolution becomes more considerable with increasing strain.25)
Accordingly, it may be expected that higher ECAP strain is effective in enhancing thermal stability of ECAPed steel. Second, dilute addition of carbide and/or nitride forming elements to steel is also very effective on increasing recrystallization temperature and on suppressing grain growth.26)

Figure 3 shows the microstructural evolution of the CS steel annealed at 813 K and 873 K for 1 h after either 4 passes or 8 passes ECAP, i.e. effective accumulated strain of 4 or 8. At 813 K, the microstructure of the 4 passes ECAPed CS steel (Fig. 3(a)) consisted of coarse recrystallized grains and fine unrecrystallized grains. By contrast, the grains of the 8 passes CS steel (Fig. 3(b)) were uniform in the size and the dislocation density remained high. At 873 K, the ferrite grains were completely recrystallized and pearlite colonies were well-defined with a dark contrast in the 4 passes ECAPed CS steel (Fig. 3(c)). However, for the 8 passes ECAPed CS steel, pearlite colonies were ill-defined (Fig. 3(d)). In addition, as shown in Fig. 4(a), the ferrite grains remained ultrafine and nano-sized particles, identified as cementite by energy dispersive spectrometer analysis (Fig. 4(b)), existed mainly at ferrite grain boundaries in the vicinity of pearlite colonies. These particles resulted from precipitation of carbon atoms dissolved from pearlitic cementite during ECAP as cementite by subsequent annealing and prevented grain boundaries from migrating by pinning effect. From the above findings, it is obvious that higher ECAP strain may enhance thermal stability in the present CS steel. It is another advantage of ECAP, at least for the steel, in addition to grain refinement since higher strain is usually known to result in faster kinetics of microstructural evolution tied to annealing.

Figure 5 presents the micrographs of the CSV steel, containing 0.06 wt% V and 0.008 wt% N, annealed at 873 K for 1 h after 4 passes ECAP. Unlike the CS steel processed by the same ECAP conditions (Fig. 3(c)), pearlite colonies were hardly defined (Fig. 5(a)) and nano-sized cementite particles were uniformly distributed throughout UFG ferrite matrix (Fig. 5(b)). It is also noticeable that dislocation densi-
ty remained high in the ferrite grains (Fig. 5(c)) and pearlitic cementite which was initially in the form of the lamellar plates became particle-like (Fig. 5(d)). This thermally stable microstructure of the CSV steel resulted from the following effects. The presence of a small amount of vanadium increases recrystallization temperature. So, the dislocation density remained high and grain boundaries were still in non equilibrium state. Dislocations and non-equilibrium boundaries provided effective diffusion path so that carbon atoms dissolved from pearlitic cementite could diffuse throughout the ferrite matrix. In the CS steel, dissolved carbon atoms could not diffuse away from pearlite colonies due to the lack of diffusion path and so the distribution of nano-sized cementite particles is restricted in the vicinity of pearlite colonies. Then, nano-sized cementite particles precipitated at the grain boundaries throughout ferrite matrix during annealing suppressed grain growth.

3.2. Tensile Properties

Figure 6 shows the room temperature nominal stress–strain curves of the CS and CSV steels before and after ECAP. Before ECAP, both steels exhibited prolonged strain hardening behavior after discontinuous yielding. The yield strength (YS) and ultimate tensile strength (UTS) of the CSV steel were higher than those of the CS steel, due to the presence of vanadium. After ECAP, the strength of both steels increased remarkably, over twice as much as before ECAP and there was no occurrence of strain hardening, as is typical for UFG materials.8,27,28) It is noted that the YS and UTS of the CS steel after 8 passes were slightly higher than those of the CS steel after 4 passes. This is attributed to the fact that the grain size after 8 passes was comparable to or slightly finer than that after 4 passes, as shown in Figs. 1(b) and 1(c). For the CSV steel, ECAP was conducted up to 4 passes, so the tensile curve for the CSV steel after 8 passes is not shown in Fig. 6. The variation of the room-temperature tensile properties of both steels with annealing temperature is plotted in Fig. 7. For the CS steel, YS and UTS of the sample annealed at 693 K are slightly less than those of the as-ECAPed sample, and decrease rapidly with further increment of annealing temperature. The loss of strength with increasing annealing temperature is attributed to two factors: the appearance of coarse recrystallized ferrite grains (see Fig. 3(a)), and softening of the pearlite due to the spheroidization of pearlitic cementite. On the contrary, the strength of the CSV steel annealed at 693 K is higher than those of the as-ECAPed sample, probably due to the precipitation of very fine Fe₃C particles. In the temperature range of 693–813 K, the strength of the CSV steel
decreased gradually with increasing annealing temperature. It is noted that the strength of the CSV steel annealed at 813 K is comparable to that of the CS steel annealed at 693 K. The extension of the mechanical stability in the CSV steel to higher temperatures resulted mainly from the preservation of a UFG ferrite grains associated with homogeneously distributed nano-sized cementite particles. The elongation to failure, $e_{f}$, of the CS steel remained unchanged up to 753 K. Then, it increased rapidly with increasing annealing temperature and finally recovered to a value close to the as-received state at 873 K. For the CSV steel, $e_{f}$ gradually increased with increasing annealing temperature. Smaller $e_{f}$ of the CSV steel than that of the CS steel above 813 K would result from its higher strength.

In Fig. 8, YS of the CS and CSV steels was plotted against $d^{-1/2}$ ($d$: ferrite grain size). In order to show the validity of the present tensile data, the YS data of UFG CS steel fabricated by warm groove rolling29) were included in the same plot. All data coalesce into a single straight line, proving the validity of Hall–Petch relation in the present ferrite grain size range of 0.2–30 μm. For the prediction of YS of low carbon–manganese steel with the ferrite grain size larger than 10 μm, the following equation30) is often quoted.

$$YS (MPa) = 15.4(3.5 + 2.1[Mn] + 5.4[Si] + 23[Ni] + 1.13d^{-1/2})$$

where [ ] is in wt%, $d$ is in mm and $N_{f}$ is the free nitrogen content. The Hall–Petch constant estimated from the slope of Fig. 8, 10.4 MPa mm$^{-1/2}$ ($=328$ MPa μm$^{-1/2}$), is lower than that in Eq. (1), $\sim 17.4$ MPa mm$^{-1/2}$. Although, at present, the origin of this difference is not clear and to be further investigated, the following postulation is worth considering. The Hall–Petch constant is regarded as a measure of resistance against yield propagation from the yielded grain to the adjacent unyielded grain. Li31) suggested that yield propagation occurs by emitting dislocations at ledges of grain boundary between the yielded grain and the adjacent unyielded grain. The large portion of grain boundaries of UFG materials fabricated by severe plastic straining remains non-equilibrium even after prolonged annealing. The Hall–Petch equation or Eq. (1) assumes that grain boundaries are equilibrium and high-angled. Accordingly, dislocation emission from non-equilibrium low-angled boundaries would be relatively easy compared to that from equilibrium high-angled grain boundaries, causing the low value of the Hall–Petch constant.

4. Conclusions

(1) An ultrafine ferrite grain structure of 0.2–0.3 μm was obtained by conducting equal channel angular pressing at 623 K to an effective strain of ~4 or ~8 in the two grades of low carbon steel, one without vanadium and the other containing vanadium, whose initial ferrite grain sizes were 30 μm and 10 μm, respectively. The as-ECAPed steels exhibited very high strength, over twice of the strength of the steels before ECAP.

(2) Ultrafine grained ferrite in the more severely pressed steel exhibited more sluggish recovery and recrystallization kinetics due to the presence of excessive carbon content in the ferrite matrix by carbon dissolution from pearlitic cementite during pressing.

(3) Under the identical annealing conditions, ultrafine ferrite grain size and the high room-temperature strength were preserved at higher annealing temperatures in the steel containing vanadium compared to the steel without vanadium. The enhanced thermal and mechanical stabilities of the steel containing vanadium were attributed to its unique microstructure consisting of ill-defined pearlite colonies and ultrafine ferrite grains with uniformly distributed nano-sized cementite particles.

(4) The yield strength of the CS and CSV steels were shown to be well fitted to the standard Hall–Petch type relationship in the ferrite grain size range of 0.2–30 μm. However, the Hall–Petch constant of the ECAPed steel was lower than that predicted from the expression developed for the yield strength of plain low carbon steels with equilibrium microstructure.
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