Ultra-fine \((\alpha+\theta)\) Duplex Structure Formed by Cold Rolling and Annealing of Pearlite

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The ferrite (\(\alpha\))-cementite (\(\theta\)) microduplex structure formed by heavy cold rolling and annealing of pearlite was studied in an Fe–1.4Cr–1.0C (mass%) alloy. Cold-rolled pearlite structure is inhomogeneous consisting of three components; (1) irregularly bent lamellae (IBL), (2) coarse lamellae with shear band (CLS) and (3) fine lamellae (FL) as was previously reported by the present authors. Misorientation in \(\alpha\) is large in the IBL and near the shear band in the CLS. As rolling reduction increases, the proportion of FL increases. By annealing at 973 K after heavy cold rolling, the \((\alpha+\theta)\) microduplex structure with \(\alpha\) and \(\theta\) grain sizes less than 0.5 \(\mu\)m is formed. This structure consists of a coarse grain region \((d_{c}>0.4 \mu m)\) containing high-angle \(\alpha\) boundaries and a fine grain region \((d_{c}=0.2 \mu m)\) with low-angle \(\alpha\) boundaries by inheriting local orientation distribution in the deformed \(\alpha\) structure. The coarse grain region is formed at the deformed region where local misorientation in \(\alpha\) is large essentially by recovery under pinning by \(\theta\) particles. As the annealing is prolonged, the fraction of the coarse grain region increases. The cold-rolled and annealed pearlite exhibits a wide range of strength-ductility balance.

KEY WORDS: microstructure; high carbon steel; pearlite; ferrite; cementite; deformation; recovery; recrystallization; grain boundary; strength; ductility.

1. Introduction

Heavily cold-drawn pearlite is the strongest structure in the steels currently used. Heavy cold deformation results in the decrease in interlamellar spacing of pearlite.\(^{1,2}\) Further, complex microstructure changes, such as nanocrystallization of cementite lamellae, dissolution of cementite into ferrite, formation of nano-ferrite and so on, occur during heavy cold drawing of pearlite.\(^{3–5}\)

The present authors studied the microstructure and mechanical property of cold-rolled pearlite.\(^{6}\) Whereas similar changes in microstructure and property by cold rolling of pearlite as in cold drawing were observed, it was found that the cold-rolled structure of pearlite was surprisingly inhomogeneous. When heavily cold-rolled pearlite is annealed, the ultra-fine grained duplex structure with ferrite (\(\alpha\)) and cementite (\(\theta\)) grain sizes less than 1 \(\mu\)m was easily obtained.\(^{7}\) However, the annealed structure was not uniform in grain size and boundary misorientation in \(\alpha\) by inheriting the heterogeneity in the cold-rolled pearlite. Recently, we studied the microstructure and mechanical property of heavily cold rolled and annealed pearlite and reported that the \((\alpha+\theta)\) microduplex structure displays a wide range of strength-ductility balance.\(^{8}\) However, the relationship between the cold-rolled structure of pearlite before annealing and the annealed \((\alpha+\theta)\) microduplex structure is not understood in detail.

The present study aims to clarify the characteristics of heavily cold-rolled pearlite focusing on the orientation distribution of \(\alpha\) in deformed pearlite and the formation process of \((\alpha+\theta)\) microduplex structure during annealing after cold rolling. Also, the tensile property of the cold-rolled and annealed pearlite is examined.

2. Experimental Procedure

A commercial high-carbon steel (SUJ2; Fe–1.4Cr–1.0C (mass%)) was used as in the previous study.\(^{7}\) Specimens, austenitized at 1 273 K for 1.8 ks, were isothermally transformed at 923 K for 0.6 ks followed by water quenching to obtain a pearlite structure. These specimens were cold-rolled in the reduction between 70% and 95% and annealed at 973 K for various periods up to 86.4 ks. Microstructure was observed by scanning electron microscope (SEM) and transmission electron microscope (TEM) along the transverse direction (TD) which is parallel to the rolling plane and perpendicular to the rolling direction (RD). Local orientation of \(\alpha\) in the deformed and annealed pearlite was determined by analyzing convergent beam Kikuchi diffraction patterns.

Tensile test specimens with the gauge size of 2.5 mm\(^{x}\)×8.6 mm\(^{y}\) were cut from the cold rolled specimens by electrode discharge machine by setting its longitudinal direction to be parallel to the rolling direction (RD), and mechanically polished to 0.5 mm thick. Tensile test was performed at an initial strain rate of 2.0×10\(^{-3}\) s\(^{-1}\).
3. Results and Discussion

3.1. Microstructure of the Cold Rolled Pearlite

Figure 1 shows the SEM microstructures of the as-transformed pearlite and the 70% cold-rolled pearlite in Fe–1.4Cr–1.0C. A lamellar pearlite with an average interlamellar spacing of 0.18 μm is formed by the isothermal holding at 923 K (Fig. 1(a)). The heavily cold-rolled structure is quite inhomogeneous as can be seen in Fig. 1(b) which was observed along three different directions of the cold-rolled specimen, i.e., ND, RD and TD.

Figure 2 shows the SEM microstructure of the specimen cold-rolled by 70%. The arrangement of lamellae is not uniform in cold-rolled pearlite. As was previously reported, the cold-rolled pearlite structure can be classified into three types shown in the corresponding schematic illustrations; (A) irregularly bent lamellae (IBL) where alignment of θ lamellae changes severely by deformation without fragmentation; (B) coarse lamellae with shear band (CLS) which consists of the area with coarse interlamellar spacing and shear bands inclined at about 30 degrees to RD; (C) fine lamellae (FL) where α and θ lamellae are almost parallel to RD with fine interlamellar spacing. Figure 3 shows the variation of deformed structure with the rolling reduction. In comparison between Figs. 3(a) and 3(b), it is clear that the fine lamella parallel to RD increases as the rolling reduction increases from 70% to 90%. A histogram of Fig. 3(c) shows change in the area fractions of the three deformed structures as a function of the reduction of cold rolling. In the 70% rolled specimen, IBL is most dominant. As rolling reduction increases, the area fraction of FL increases and exceeds 60% after 95% cold rolling.

Figure 4(a) is the TEM micrograph of IBL in the specimen rolled by 70% on which local misorientation in α is described. The orientation of α changes largely across θ lamellae. Also, there are many cell boundaries within the same α lamella. Especially, the cell boundaries where the lamellae are severely bent exhibit large misorientations. This kind of large change in local orientation is similar to those of “kink band” or “matrix band” observed in the deformed single-phase metallic materials. Fig. 4(b) is the TEM micrograph of CLS in the specimen cold-rolled by 90%. The interlamellar spacing of the coarse lamella area is larger than 0.1 μm which is not reduced much from the initial spacing. In contrast, the interlamellar spacing in the shear band is reduced significantly from the initial spacing,
implying that the deformation is localized at the shear band in the CLS region. Misorientation between the adjacent ferrite lamellae is small (less than 15 degrees in Fig. 4(b)) in the coarse lamellar area. In contrast, the orientation of a in the shear band is largely different from that in the coarse lamellar area.

Figures 5(a) and 5(b) show the TEM microstructure of FL in the specimen rolled by 95% and the illustrations showing misorientation between adjacent lamellae. Interlamellar spacing is reduced to 20 nm. Each a lamella corresponds to the a lamella in the original pearlite and is bounded by $\theta$ lamellae. The misorientation between adjacent $\alpha$ lamellae across the $\theta$ lamella is relatively large and often reaches to 10 degrees as described in Fig. 5(a) although this misorientation does not accumulate macroscopically as seen in the point-to-origin misorientation in Fig. 5(b). It is considered that each $\alpha$ lamella is alike an elongated dislocation cell. Since the cell width (i.e., the width of $\alpha$ lamella) is extremely smaller than that of deformed $\alpha$ single-phase typically 0.3 $\mu$m, it is considered that the $\theta$ lamella bounded the $\alpha$ lamellae is a strong barrier against slip in $\alpha$ and geometrically necessary dislocations are accumulated at the $\alpha$/$\theta$ boundary resulting in the relatively large misorientation.

3.2. Microstructure Change of the Cold Rolled Pearlite by Annealing

Figure 6 shows the TEM microstructure in the early stage of annealing of the IBL area in the cold-rolled pearlite. The specimen was annealed at 973 K for 0 s, i.e., immediately quenched after the temperature reached to 973 K. In Fig. 6(a), the $\theta$ lamellae undergo spheroidization and simultaneously, formation of equi-axed fine $\alpha$ grains is observed. However, such a microstructure change proceeds quite heterogeneously. Formation of the $(\alpha+\theta)$ microduplex structure consisting of equi-axed $\alpha$ grains and spheroidized $\theta$ particles is initiated at the area where lamellae are heavily bended in Fig. 6(b) which is an enlargement of the area 1 in Fig. 6(a). Figures 6(c) and 6(d) are an enlargement of the area 2 in Fig. 6(a) and the corresponding map showing the $\alpha$ boundary misorientations, respectively. In the upper portion of Fig. 6(c) where a coarser lamellar structure is remained due to relatively low degree of local deformation, $\theta$ lamellae are not spheroidized much and subgrain boundaries are contained in the $\alpha$ lamellae. Equi-axed $\alpha$ grains, mostly surrounded by high-angle boundaries and spheroidized $\theta$ particles are seen in the lower portion of Fig. 6(c).

Figures 7(a) and 7(b) are TEM micrographs showing the early stage of annealing of CLS and FL areas, respectively. In the CLS (Fig. 7(a)), $\theta$ phase in the coarse lamellar area remains unspheroidized but fine microduplex structure is formed along the shear band. In the FL (Fig. 7(b)), $\theta$ is disconnected by spheroidization but still elongates on the ND plane. The deformed $\alpha$ still keeps the lamellar-like cell morphology as in the cold-rolled specimens although the cell width (in another word, the interlamellar spacing) is increased by about two times due to recovery after the short annealing treatment employed. The presence of the misorientation between adjacent $\alpha$ lamellae in Fig. 5(a) is more evident since $\alpha$ subgrain boundaries are clearly seen as pointed by an arrow. Thus, it is concluded that the formation of equi-axed $(\alpha+\theta)$ microduplex structure occurs in the deformed region with...
large α misorientation in the IBL and CLS regions.

After further annealing, the (α + θ) microduplex structure is formed throughout the specimens but is still not uniform in α and θ grain sizes. Figure 8(a) is the SEM micrograph of the specimen annealed at 973 K for 18 ks after 70% cold rolling. The annealed structure consists of two regions; (1) coarse grain region larger in α grain size and θ particle size and (2) fine grain region with smaller α grains and θ particles. Figures 8(b) and 8(c) show that the fraction

Fig. 6. TEM microstructures (TD plane) of IBL in the specimen annealed at 973 K for 0 s after 70% cold rolling in Fe−1.4Cr−1.0C; (a) a low-magnification image, (b) an enlargement of the area 1 in (a), (c) an enlargement of the area 2 in (a) and (d) the corresponding map showing α boundary misorientations.

Fig. 7. TEM microstructures (TD plane) of the pearlite specimen annealed at 973 K for 0 s after cold rolling in Fe−1.4Cr−1.0C; (a) CLS, cold-rolled by 70% and (b) FL, cold-rolled by 95%.

Fig. 8. (a) SEM micrograph (TD plane) of the pearlite cold-rolled by 90% and annealed at 973 K for 0.6 ks in Fe−1.4Cr−1.0C, (b), (c) the changes of area fraction of the microstructure components during annealing at 973 K after 70% and 90% cold rolling, respectively.
of coarse grain region increases whereas that of fine grain region decreases. Also, a small fraction of lamellar region is contained in the specimen annealed after 70% cold rolling in Fig. 8(b). By increasing the rolling reduction to 90%, such lamellar region is eliminated after annealing (Fig. 8(c)).

Figure 9 shows the TEM micrograph and corresponding illustrations showing misorientation across \( \alpha \) boundary for those two regions in the specimen annealed at 973 K for 120 s after 90% cold rolling. The coarse grain region \((d_{\alpha}\sim 0.4 \mu m, d_{\gamma}\sim 0.2 \mu m)\) in Fig. 9(b) contains many \( \alpha \) grain boundaries with large misorientation. On the other hand, most of \( \alpha \) grains are subgrains surrounded by low-angle boundaries in the fine grain region \((d_{\alpha}\sim 0.2 \mu m, d_{\gamma}\sim 0.1 \mu m)\) in Fig. 9(c). Since Figs. 6 and 7 have shown that the spheroidization of \( \gamma \) and the formation of equi-axed \( \alpha \) grains surrounded by high-angle boundaries start at IBL and at the shear band of CLS, it is concluded that those areas with large misorientation in the deformed \( \alpha \) grain turn to be the coarse grain regions with high-angle \( \alpha \) boundaries whereas the fine grain region containing \( \alpha \) subgrains originates from FL and coarse lamella areas in IBL and CLS.

The equi-axed \( \alpha \) grains surrounded by high-angle boundaries appear to be formed by recrystallization. Ordinarily, characteristic textures are obtained after the cold rolling as well as after the recrystallization of \( \alpha \). Figure 10 shows the \([200]_{\alpha}\) pole figures of the cold-rolled and annealed specimens. As is seen in Figs. 10(a) and 10(b), typical rolling textures of which components are \(\{112\}(110), \{111\}(112)\) and \(\{001\}(110)\) are obtained by heavy cold rolling. As the rolling reduction increases from 70% (Fig. 10(a)) to 90% (Fig. 10(b)), this rolling texture seems to be enhanced. After the annealing at 973 K for 1.8 ks, there is no significant change in the texture component except the increase in the sharpness of the rolling texture implying that only recovery takes place in the deformed \( \alpha \). The \( \alpha \) grains surrounded by high-angle boundaries in Figs. 6(a) and 6(b) are recrystallized \( \alpha \) grains. However, they can grow hardly by the pinning effect of fine \( \gamma \) particles since the \( \theta \) volume fraction is high (ca. 15%) in the high-carbon steel used. As were seen in Figs. 8(b) and 8(c), gradual increase in the area fraction of the coarse grain region occurs during the prolonged annealing according to the slow \( \theta \) coarsening in the Cr-contained alloy used.

Figure 11 schematically describes the formation process of the \((\alpha + \gamma)\) microduplex structure by the annealing of the cold-rolled pearlite. The annealed structure inherits the heterogeneity in the cold-rolled pearlite structure. A part of the IBL region and the shear bands of the CLS region exhibit large local misorientation in the \( \alpha \) matrix. After annealing, the spheroidization of \( \gamma \) occurs and equi-axed \( \alpha \) grains surrounded by high-angle boundaries are formed in those regions with large misorientation, resulting in the coarse
grain region with high-angle $\alpha$ boundaries. Since finely dispersed $\theta$ particles provide large pinning effect, no appreciable migration of grain boundary occur. This leads to the formation of the equi-axed $\alpha$ grains essentially by recovery. This phenomena can be understood as ‘in-situ’ or ‘continuous’ recrystallization\(^2\) which does not accompany the significant migration of high-angle boundaries. Local misorientation in $\alpha$ is small and the spheroidization of the $\theta$ lamellae is slow due to rather small deformation in the coarse lamellar region of the IBL and CLS. So these structures turn to be the fine grain region containing low-angle $\alpha$ boundaries. The FL region is largely but uniformly deformed so that the misorientation between adjacent $\alpha$ lamellae is not large. Since the spacing of the $\theta$ lamellae is small, the fine $\theta$ dispersion is obtained after the spheroidization. The fine grain region with $\alpha$ subgrains is finally developed in the FL region by recovery. Those fine grain regions are gradually expelled by the grain growth of $\alpha$ grains in the coarse grain region according to the $\theta$ coarsening during the further annealing.

3.3. Mechanical Property of the Cold-rolled and Annealed Pearlite

Figures 12(a) and 12(b) show the yield strength–ductility (total elongation) and tensile strength–ductility balances of the as-transformed pearlite, the cold-rolled pearlite and the ($\alpha + \theta$) microduplex structures formed by the annealing after the cold rolling. Figure 12(c) is the corresponding stress–strain curves. The as-transformed pearlite is relatively low in yield strength (0.2% proof strength is 754 MPa) but exhibits large work hardening resulting in tensile strength of 1 220 MPa and total elongation of about 10%. After 90% cold rolling, the yield strength of deformed pearlite is increased by more than twice up to 1 871 MPa. It is known that the strength increases exponentially as strain increases in cold drawing.\(^2\) This large increase of strength is consistent with the previous study on heavy cold rolling of carbon steels.\(^4\) The increase of the rolling reduction from 70% to 90% does not change ductility but increases...
strength. Large work hardening is also seen during the tensile test of the heavily cold-rolled pearlite, resulting in the high tensile strength of 2220 MPa in the 90% cold-rolled specimen. This is presumably due to the lamellar morphology of $\theta$ even though it was fragmented or changed partly to nano-sized crystals after severe plastic deformation.\footnote{15,16} A superior strength–ductility balance was also obtained in the pearlite cold-rolled and annealed at a lower temperature for a shorter period, which maintain the near-lamellar $\theta$ morphology.\footnote{81} By increasing the annealing period at 973 K, strength decreases and ductility i.e., total elongation increases despite of decrease in work hardening seen in Fig. 12(c). Sherby and his co-authors\footnote{13,14} also reported that the $(\alpha + \theta)$ microduplex structure in ultra-high carbon steels exhibits high strength and good ductility at room temperature.

Fine-grained $\alpha$ of submicron grain size in pure iron or interstitial-free steels exhibits high strength over 800 MPa but exhibit poor uniform elongation due to lack of work hardening.\footnote{15,16} The presence of hard second phase ($\theta$ or martensite) enhances the work hardening of $\alpha$ which results in the increase of uniform elongation and total elongation in low carbon steels.\footnote{15} In high-carbon steels, Onel and Nutting\footnote{15} showed that the work hardening increases as the $\theta$ volume fraction increases in the tempered martensite. It is considered that the superior ductility of the $(\alpha + \theta)$ microduplex structure in the present study compared with the fine-grained $\alpha$ single-phase structure, e.g., the total elongation of 15% and the yield strength of 950 MPa of the 90% cold-rolled and annealed at 973 K for 1.8 ks, is mainly due to the presence of fine $\theta$ particles. In ultra-high carbon steels, however, Syn et al.\footnote{18} reported that tensile ductility is limited since the fracture strength is affected by the size of coarse carbide and increases as the work hardening decreases. Those effects by carbide are contradicting each other and thus, need to be compromised to obtain the best strength–ductility balance. Further study is necessary to understand the microstructure–property relationship of high-carbon steels.

### 4. Conclusion

Ultra-fine (2) grained $(\alpha + \theta)$ duplex structure with $\alpha$ and $\theta$ grain sizes less than 0.5 $\mu$m is obtained in the Fe–1.4Cr–1.0C alloy by heavy cold rolling and annealing of pearlite structure. The following conclusions were drawn.

(1) Cold-rolled pearlite structure is inhomogeneous depending on the initial alignment of $\theta$ lamellae in pearlite colony. As rolling reduction increases, the proportion of the area with fine interlamellar spacing increases.

(2) By annealing at 973 K after heavy cold rolling, the $(\alpha + \theta)$ microduplex structure with $\alpha$ and $\theta$ grain sizes less than 0.5 $\mu$m is formed. This structure consists of the coarse grain region containing high-angle $\alpha$ boundaries and the fine grain region with low-angle $\alpha$ boundaries by inheriting the local orientation distribution of $\alpha$ in the deformed structure. Such microduplex structures are formed essentially by recovery due to the strong pinning effect of $\theta$ with a high volume fraction.

(3) The cold-rolled and annealed pearlite exhibits a wide range of strength–ductility balance. The fine dispersion of $\theta$ provides reasonable uniform elongation in the ultra-fine grained $\alpha$ steel.

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### REFERENCES