Temperature Dependence of Grain Boundary Structure and Grain Growth in Bulk Silicon–Iron

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Grain boundary shapes and grain growth in bulk 2.61 wt% silicon–iron have been studied by heat-treating at temperatures between 700 and 1 200°C. Initial microstructure with fairly uniform fine grains has been obtained by recrystallization at 800°C for 5 min after deformation. When subsequently heat-treated at 700 and 800°C, a fraction of the grain boundaries have hill-and-valley shapes with several facet planes or kinks. Some of these facet boundary segments are expected to be singular. Abnormal grain growth occurs at 700 and 800°C and is attributed to step growth of the boundaries. When heat-treated at 1 000°C, all grain boundaries are defaceted with smoothly curved shapes, indicating that they are atomically rough. At temperatures above 1 000°C, normal grain growth occurs, because the rough grain boundaries move continuously. This correlation between grain boundary structure and grain growth is consistent with the earlier observations in other metals and oxides. It is thus shown that the abnormal grain growth in this alloy occurs at low temperatures because of the singular grain boundary structure.

KEY WORDS: silicon–iron; silicon–steel; abnormal grain growth; grain boundary faceting; singular grain boundary.

1. Introduction

In production of Si–steel sheet, the \{110\}<001> Goss texture develops by abnormal grain growth (AGG) (also referred to as secondary recrystallization). Therefore, the mechanism of AGG has been extensively studied in this alloy. It has been proposed, based on experiments, that this phenomenon arose from the grain boundary pinning by precipitates,1–3) surface energy anisotropy and surface grooving,4–7) recrystallization texture, 8,9) or grains with coincidence site lattice (CSL) orientations. 10–12) But the dominant mechanism is yet to be determined. One of the unresolved questions is if AGG can occur in this alloy without developing any texture.

It was recently observed that AGG and normal grain growth (NGG) in bulk polycrystalline metals 13–18) and oxides19–20) were related to grain boundary structures. At low temperatures or in the presence of additives, grain boundaries had hill-and-valley (h&v) shapes with singular segments and AGG occurred. At high temperatures or with different additives, all grain boundaries were defaceted and NGG was observed.13,14,16–20) The occurrence of AGG was attributed to the step growth of singular grain boundaries.21–26)

The purpose of this work is to determine experimentally if the same correlation between the grain growth and grain boundary structure exists in pure Si (about 3 wt%) iron at different heat-treatment temperatures. By using bulk polycrystalline specimens, the effects arising from surface and texture can be eliminated. The previous observations in pure and single phase metals show that grain boundaries can undergo roughening transitions in the temperature range of 0.6–0.9 \(T_m\), where \(T_m\) is the melting point in absolute scale.13–18,27–29) For Si–iron, this will be about 780–1 300°C. Previous evidences for possible grain boundary roughening transition in pure Si–iron can be found in the grain boundary migration experiments with bicrystals by Tsurekawa et al.30)
ing along the grain boundaries using an image analysis program. For the specimens with fine grains, about 500–2,000 grains were measured, and for those with large grains, about 100–300 grains were measured. The grain boundary shapes were also examined in a transmission electron microscope (TEM).

3. Results and Discussion

Wet chemical analysis of the heat-treated bar showed that the Si content was 2.61 wt% and the major impurities were P (0.015 wt%), Ni (0.06 wt%), and Cr (0.01 wt%). The C, Mn, and S contents were about 0.005 wt%. The specimens heat-treated at 800°C for 5 min after compressing to 66.7% in height had typical primary recrystallization structures with fine grains of 8.7 μm in average radius as shown in Fig. 1. The X-ray pole figure and electron back-scattered pattern (EBSP) analysis of these specimens did not show any macroscopic or microscopic texture. Figure 1 is the initial microstructure for all subsequent heat-treatments at various temperatures to observe the grain growth behavior.

The grain growth during the heat-treatment at 600°C was very slow. After 250 h at 600°C, the grains were only slightly larger than those shown in Fig. 1. During the heat-treatment at 700°C, typical AGG behavior was observed as shown in the microstructures of Fig. 2, and grain size distributions in Fig. 3. After 3 h, some large grains began to appear as shown in Fig. 2(a), and after 24 h, some grains had grown to about 700 μm radius as shown in Fig. 2(d). In Fig. 3, those large grains that could be clearly identified as the abnormal grains are marked by arrows. The fine matrix grains grew slowly as can be seen in Figs. 2 and 3.

When the heat-treatment temperature was increased to 800°C, distinct AGG behavior was again observed during 24 h, but the number density of the abnormal grains was larger than that observed at 700°C as shown in Fig. 4(a) for an intermediate growth stage. After 24 h, a few grains grew to sizes as large as 600 μm radius. The large grains began to appear after 30 min (compared to 3 h at 700°C) and the matrix grains also grew faster than at 700°C. The grain growth during the heat-treatment at 900°C for periods up to 70 h appeared to be nearly normal as shown in Fig. 4(b) at an intermediate growth stage. Although the normalized (to the average size) grain size distributions were narrow and appeared to be constant at increasing heat-treatment periods, there were a small fraction of grains 5 to 7 times larger than the average size. Such large grains were not observed during the normal growth at 1,000, 1,100, and 1,200°C as

![Fig. 1. The optical microstructure of the specimen heat-treated at 800°C for 5 min after compressing to 66.7% representing the initial grain structure for all subsequent heat-treatments.](image)

![Fig. 2. The optical microstructures of the specimens heat-treated at 700°C for (a) 3 h, (b) 6 h, (c) 12 h, and (d) 24 h.](image)
will be shortly described. Therefore, it appeared that there still was a slight tendency for AGG at 900°C.

Normal grain growth was observed during the heat-treatment at 1000°C as shown in Fig. 5 for selected periods. The grain size distributions normalized to the average value were narrow and did not change with the heat-treatment time as shown in Fig. 6. When the heat-treatment temperature was increased to 1100 and 1200°C, NGG was again observed with the invariant normalized grain size distributions resembling those shown in Fig. 6. The grain growth rate increased with temperature as expected.

The grain boundaries in the selected specimens heat-treated at various temperatures were examined under TEM and their typical shapes are shown in Figs. 7 and 8. The observed shapes of grain boundaries could be classified into three types. The first type (shown in Fig. 7(a)) had multi-faceted h&v shapes with several alternating facet planes. The second type (shown in Fig. 7(b)) showed two grain boundary segments meeting at a kink. In the third type (shown in Fig. 8), the grain boundary appeared to be smoothly curved. In the specimens heat-treated at 700 and 800°C, all of these grain boundary types were observed. In Fig. 7(a), for example, a multi-faceted h&v grain boundary met two grain boundaries at a triple junction (indicated by an arrow), which were observed to be smoothly curved. In Fig. 7(b), a grain boundary with a kink met two curved grain boundaries at the triple junction (indicated by an arrow). Some grain boundaries had combinations of multi-faceted h&v, kinked segments, and smoothly curved segments. Out of about 110 grain boundaries observed in the specimen heat-treated at 700°C for 24 h (shown in Fig. 2(d)), 8 had multi-faceted h&v segments and 15 had kinks. The rest were smoothly curved. Out of about 120 grain boundaries observed in the specimen heat-treated at 800°C for 30 min, 4 had multi-faceted h&v segments and 12 had kinks. The rest were smoothly curved. All of about 60 grain boundaries examined in the specimen heat-treated at 1000°C for 1.5 min (shown in Fig. 5(b)) were smoothly curved as shown in Fig. 8.

As pointed out earlier,13–20,31,32) some flat segments of the h&v grain boundaries are likely to be singular corresponding to the cusps in the grain boundary Wulff plot.33) The kinked shapes are the limiting cases of the h&v shapes, because the h&v shapes can evolve to the kinked shapes by a coarsening process. Therefore, some flat grain boundary segments meeting other segments at a kink are also likely to be singular. It is well known34) that the surfaces of polycrystalline specimens often show thermal faceting with h&v shapes. Herring35) showed that if the equilibrium shape of a crystal has sharp edges or corners where the surface normal changes discontinuously, a surface with an average orientation that does not appear in the equilibrium will have an h&v shape. The equilibrium shapes of crystals at moderately high temperature will have flat singular segments and curved edges. The curved segments are atomically rough as shown by Herring,35) Rottman and Wortis,36) and Jayaprakash and Saam.37) Therefore, at least some segments...
of the h&v surface are likely to be singular while the others could be rough. With temperature increase, the relative area of the rough segments will increase until the entire h&v surface becomes defaceted and rough. The defaceting transition is thus a manifestation of the roughening transition at the edges and corners of an equilibrium shape. There is the same relationship between the equilibrium shapes of grains embedded in other grains and h&v shapes of their grain boundaries. Therefore, when these h&v grain boundaries become defaceted at 1000°C to smoothly curved shapes, they become atomically rough. If a singular grain boundary is rapidly moving, its shape may become curved by kinetic

Fig. 5. The optical microstructures of the specimens heat-treated at 1000°C for (a) 1 min, (b) 1.5 min, (c) 2 min, and (d) 15 min.

Fig. 6. The measured grain size distributions of the specimens normalized to the average value heat-treated at 1000°C for various periods.

Fig. 7. The TEM micrographs of faceted grain boundaries (a) with hill-and-valley structure in the specimen heat-treated at 700°C for 24 h and (b) with a kink in the specimen heat-treated at 800°C for 30 min.
roughening as a surface. It is therefore possible that not all singular grain boundaries are identified by their shapes in the specimens heat-treated at low temperatures. The grain boundary defaceting transition observed in this Si–iron is similar to those observed in other metals. The transition temperature of approximately 1 000°C (about 0.73 Tm) also falls into the range of 0.6–0.9 Tm observed in other metals, although the transition temperatures vary among the grain boundaries and depend also on additives and impurities. In 316L stainless steel, the grain boundary defaceting transitions were observed at temperatures between 1150 and 1200°C. As briefly mentioned in Introduction, Tsurekawa et al. examined grain boundary migration in bicrystals of 3.28 wt% Si–iron specimens. Their microstructures show kinks in both (221) 29 and random boundaries when heat-treated at 997°C. At this temperature, the migration rate, v, was low at low driving forces and increased linearly with it after abruptly increasing at a certain value. At 1 047°C, v increased still non-linearly with driving force, F, but the abrupt increase occurred at a lower F than at 997°C. Such a non-linear v(F) relationship is consistent with the step growth mechanism of singular grain boundaries proposed by Gleiter, Babcock and Balluffi, and Rae and Smith. The observed difference between the two temperatures is also consistent with the decrease of the grain boundary step free energy associated with the roughening at increasing temperatures. Tsurekawa et al. observed furthermore that the grain boundary mobilities increased abruptly in the temperature range between about 1 000 and 1 100°C. This result is also consistent with the grain boundary roughening transition, and the transition temperature range is close to about 1 000°C observed in this experiment. Although Tsurekawa et al. interpreted their results in terms of segregation effect (because they apparently thought that the non-linear v(F) was inconsistent with singular grain boundaries), their results, specially with the kinked grain boundary shapes, appear to be more consistent with the grain boundary roughening transition. The results of Tsurekawa et al. and this experiment thus provide fairly consistent evidences for the grain boundary roughening transition in this alloy.

Hall et al. observed that [110] tilt grain boundaries in Fe–3wt%Si sheet specimens annealed at 1 050–1 200°C showed h&v structures at fine scales. It is likely that these grain boundaries were actually rough with curved shapes at the annealing temperatures and the fine scale h&v structures formed during cooling. If the grain boundaries undergo roughening-singular transitions, their structures at low temperatures will depend on the cooling rate.

The correlation between the grain boundary roughening transition and the change of grain growth mode from AGG to NGG is also consistent with the previous observations in metals and oxides. As proposed earlier, if the singular grain boundaries migrate by the movement of existing steps or those produced by two dimensional nucleation, v(F) will be non-linear as observed by Tsukawaka et al. in polycrystals with singular grain boundaries, non-linear v(F) will cause AGG as confirmed by simulation. When the singular boundaries coexist with rough ones, they can still control the overall growth behavior and hence cause AGG. When all grain boundaries become rough, they will undergo continuous migration with linear v(F). Then NGG will occur. Therefore, the change of the grain growth behavior from AGG to NGG is a manifestation of the grain boundary roughening transition.

The results of this work and those in 316L stainless steel show that the transition from AGG to NGG with temperature increase occurs gradually, and at intermediate temperatures (as, for example, 900°C for this alloy) the classification to either AGG or NGG can be ambiguous. Such observations are, however, consistent with gradual decrease of the grain boundary step free energy with temperature as predicted and observed for surface grain boundaries and the transition temperatures varying among the grain boundaries.

The grain growth in this alloy at temperatures above 1 000°C was determined to be NGG on the basis of invariant normalized grain size distributions. The average grain size increased as square root of heat-treatment time at 1 100 and 1 200°C, but at 1 000°C, the average grain size after a long heat-treatment was lower than that expected from this growth law. Such a slow down of grain growth was attributed to solute segregation at grain boundaries.

May and Turnbull suggested that NGG occurred at all temperatures in Si–iron sheet on the basis of heat-treatment for 15 min. The results of this work indicate that this heat-treatment time was probably too short to observe AGG at low temperatures. The grain growth behavior can be completely determined, as shown in this work, by making observations at varying heat-treatment times.

In commercial Si–steel sheets, there can be additional effects arising from precipitates, surface energy anisotropy, and texture. The theoretical models for these effects also assume non-linear v(F) arising from the grain size dependent effective mobility. An intriguing question is if AGG will be caused by precipitates, surface energy anisotropy, or texture even when all grain boundaries are rough. Such experiments have not yet been done.

Finally, in order to check the possible effect of CSL grain boundaries on AGG of this alloy, the EBSP of the rectangu-
Fig. 9. The EBSP image of the rectangular region in Fig. 2(d) with the low $\Sigma$ boundaries shown as white lines.

lar region between a large abnormal grain and many fine grains marked in Fig. 2(d) was obtained as shown in Fig. 7. Those grain boundaries with $\Sigma$ values less than 27 were identified by using the Brandon criterion$^{56}$ and are exhibited as white lines. This result shows that although there are some low $\Sigma$ boundaries between the abnormal grain and the fine grains, they are not more numerous than those between the fine grains. Therefore, the CSL grain boundaries do not seem to be important for AGG of this alloy.

4. Conclusions

This work has shown that a fraction of grain boundaries in bulk 2.61 wt% silicon iron have h&v shapes, indicating that some flat segments are singular at 700 and 800°C. At these temperatures, AGG occurred. At 1000°C and above, all grain boundaries became deflected with smoothly curved shapes, indicating rough structures. Normal grain growth occurred at these temperatures. This correlation between the grain boundary structure and grain growth is similar to those found in other metals$^{13-18}$ and oxides.$^{19,20}$ The results of this study thus confirm that AGG can arise from the singular grain boundary structure without any precipitates, surface energy anisotropy, or texture. The normal grain growth can be characterized by invariant normalized grain size distribution. These observations are contrary to the earlier observations of May and Turnbull$^3$ which showed that NGG occurred at all temperatures in an alloy of similar composition.

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