1-1-2002

Delta-Ferrite Recovery Structures in Low Carbon Steels

Rian J. Dippenaar
University of Wollongong, rian@uow.edu.au

Dominic J. Phelan
University of Wollongong, phelan@uow.edu.au

Delta-Ferrite Recovery Structures in Low Carbon Steels

R. J. DIPPENAAR and D. J. PHELAN
BHP Institute for Steel Processing and Products, University of Wollongong

The development of delta-ferrite recovery sub-structures in low carbon steels has been observed in-situ utilising laser scanning confocal microscopy (LSCM). Well developed sub-boundaries with interfacial energies much smaller than that of delta-ferrite grain boundaries formed following transformation from austenite to delta-ferrite on heating. Transformation stresses associated with the austenite to delta-ferrite phase transformation generate dislocations that subsequently recover into sub-boundaries by a process of polygonisation. Experimental evidence in support of this proposal was found in a ferritic stainless steel. Thermal cycling through the high temperature delta-ferrite/austenite/delta-ferrite phase transformation leads to the development of a strong recovery substructure, which in turn, modifies the low temperature austenite decomposition product from Widmanstätten/bainite to polygonal ferrite, with a commensurate change in hardness.

INTRODUCTION

ALTHOUGH it is generally conceded that the early stages of solidification and subsequent high-temperature phase transformations profoundly influence cast structure, conclusions have mostly been drawn from indirect experiments and very little work has been done on the direct observation of events. Of special interest is a fundamental understanding of the events occurring in the meniscus region of high-speed continuous casters that determine the quality of the cast product. The delta-ferrite to austenite phase transformation occurs when the newly formed steel shell is relatively thin. The volume change and differences in thermal expansion of the phases may generate stresses, which, if the strength of this thin shell is exceeded, can lead to casting defects. Moreover, the delta-ferrite to austenite phase transformation may also play a role in the subsequent decomposition of austenite and through this, the microstructural development on further cooling and hence, the mechanical properties of the final product. An impediment to success in previous studies has been the inability to study this high temperature transformation directly because subsequent phase transformations mask the transformation mode.

The final alpha-ferrite grain size following decomposition from austenite in plain carbon steel is largely controlled by the grain size of the parent austenite as austenite grain boundaries and in particular grain corners, are the preferred sites for the nucleation of alpha ferrite[1,2,3,4]. Therefore, a smaller austenite grain size will lead to refinement of the alpha ferrite grain size. In strip casting, and to a lesser extent in thin-slab casting, the opportunities for control of the microstructure through thermo-mechanical processing[5] (TMP) are restricted. Therefore the exact way in which the delta-ferrite to austenite phase transformation occurs following solidification becomes increasingly important[6]. The occurrence of subsequent transformations at lower temperature masks the prior delta ferrite transformation and hence quench–arrest techniques cannot be used. A second obstacle to a detailed study of delta-ferrite decomposition is the difficulty in obtaining sufficient resolution at the high temperatures pertaining to this phase transition; the emission of infrared light from a sample at such high temperatures renders the in-situ observation of events by optical microscopy very difficult.

The phase transformation from austenite to ferrite is accompanied by the generation of dislocations, which through a subsequent process of recovery, can be rearranged into sub-

boundaries, one such example being veining[7]. Hauser[8] have shown that transformation stresses accompanying phase transformations can produce dislocation networks, even in the absence of mechanical working and this phenomenon has been observed in alpha ferrite and a number of other systems[9,10]. At elevated temperature these dislocations can be reorganised into sub-grain boundaries through a process of recovery, or more specifically polygonisation. Hence, if austenite is heated into the delta-ferrite region, it might be expected that the transformation stresses accompanying this phase transition may result in dislocation structures, which at the high pertaining temperatures, could form sub-grain boundaries through polygonisation. Moreover, Furuhara and Maki[11] have recently shown that sub-boundaries can also act as sites for austenite nucleation in duplex stainless steels. It therefore seems probable that the formation of a recovery structure in delta-ferrite could offer a means of microstructural control in the absence of mechanical deformation. Cycling through the austenite to delta-ferrite phase transformation could lead to the generation of recovery structures. If these structures were to increase the number of sites available for austenite nucleation on cooling, the grain size of the austenite would be refined and subsequently, the number of sites for alpha ferrite nucleation would be increased, leading to a more refined final alpha-ferrite microstructure.

EXPERIMENTAL

High temperature laser-scanning confocal microscopy (LSCM)

In confocal microscopy, laser light is focused by an objective lens onto the object and the reflected beam is focused onto a photo detector via a beam splitter. An image is built up by scanning the focussed spot relative to the object, which is then stored in an imaging system for subsequent display. Through the use of a confocal pinhole, only light incident from the focal plane is permitted to pass through to the photo detector. Hence, an extremely thin optical section is created, providing a sharp image at high resolution. Because thermal radiation is also blocked by the confocal pinhole, only the polarised reflection of the high intensity laser beam reaches the imaging sensor and a sharp image is produced. In these experiments, magnifications up to 1350x were used. The laser beam, a He-Ne laser with a wavelength of 632.8nm and 0.5μm diameter, is reflected and scanned by an acoustic optical deflector in the horizontal direction at a rate of 15.7kHz and a galvano-mirror in the vertical direction at 60Hz. Specimens are placed at the focal point of a gold-plated ellipsoidal cavity in an infrared furnace beneath a quartz view port, as shown in Figure 1. A 1.5kW-halogen lamp located at the other focal point in the cavity heats the specimen by radiation. The specimen and lamp chambers are separated by quartz glass so that the atmosphere of the specimen chamber can be controlled and the lamp air-cooled.

A schematic diagram of the infrared furnace and a specimen holder is shown in Figure 1.

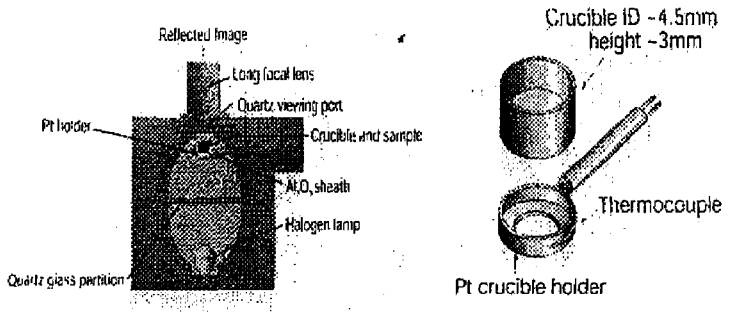


FIGURE 1 Schematic diagram of the infrared furnace and holders

Generation of sub-boundaries in delta-ferrite

The delta-to-gamma and reverse phase transition was studied in two commercial low alloy steels containing 0.06 percent carbon by mass. Table 1. The samples were heated to 1400°C (γ-phase) and held for 10 minutes, before heating at a rate of 100°C/min to 1450°C (δ-phase). Samples were then held at this temperature. This heat treatment rendered a sub-grain structure in delta-ferrite. In an attempt to prove that the phase transformation from austenite to delta-ferrite generates dislocations, which subsequently arrange themselves into low angle boundaries through recovery, a series of experiments were conducted on a steel commercially known as 3CR12, Table 1. A two-phase region, austenite plus delta-ferrite exists between 1000°C and 1150°C in this steel. Soaking at 1400°C for 10 minutes allowed sufficient time for large stable delta-ferrite grains to form. The sample was then cooled at 100°C/min to 1100°C (within the two-phase region), and sufficient time allowed for the phases to reach equilibrium. The sample was then reheated to 1400°C while the transformation of austenite to delta-ferrite was continuously observed and the development of the microstructure recorded.

TABLE 1 Composition of alloys (mass%)

Steel	C	P	Mn	Si	Al	S	Cr
Al killed	0.06	0.11	0.23	<0.005	0.04	0.014	
Si killed	0.06	0.10	0.40	0.29	<0.005	0.014	
3CR12 (typical)	0.03	0.02	1.20	0.40		0.005	11.36

Thermal cycling through the delta/gamma phase transition

The low carbon, silicon-killed and aluminium-killed steels listed in Table 1 were heated at 100°C/min to 1450°C and held for 10 minutes so that large stable grains of delta-ferrite were obtained. The specimens were then cycled between 1450°C and 1350°C, the single-phase regions of delta-ferrite and austenite respectively, at a heating and cooling rate of 300°C/min. The hold time at 1450°C and 1350°C was varied between 0 and 60 seconds. The specimens were then cooled at 100°C/min to 700°C and held until austenite decomposition was complete. The details of the cycles for each experiment are shown in Table 2.

TABLE 2 Details of thermal cycles for silicon and aluminium killed steel

	Test 1	Test 2	Test 3	Test 4	Test 5
Hold time at 1450°C (seconds)	0	10	20	30	60
Hold time at 1350°C (seconds)	0	10	20	30	60
Number of cycles	6	6	6	6	6

RESULTS

Interpretation of laser-scanning confocal microscopy observations

It is important to reflect upon the interpretation of in-situ observations made in the LSCM. The volume change accompanying a phase transformation leads to a raising or lowering of the specimen surface, locally associated with the moving interface and this leads to a change in contrast. Therefore thermal cycling leading to repeated transformations will result in roughening of the surface to the point where the image becomes diffuse. On the other hand, surface diffusion acts to smooth the surface especially at high temperature.

High-energy grain boundaries leads to grooving of the surface as a result of diffusion of atoms from the line defect to the surface, resulting in a profile such as that presented schematically in Figure 2. The existence of such a 'V'-groove alters the optical path of the reflected light, some reflected rays are scattered, leading to the development of contrast. The formation of ridges on the free surface where line defects or other interfaces intersect the

surface is a result of diffusion along the interface to the free surface being faster than diffusion away from the ridged area along the surface[12]. Because a ridge such as shown in Figure 2, is likely to form whenever grain or phase boundaries intersect the free surface, the appearance of all such boundaries in LSCM should be similar: a dark line boarded by areas of light contrast. Examples of different kinds of boundaries are shown in Figure 2 (a) and (b).

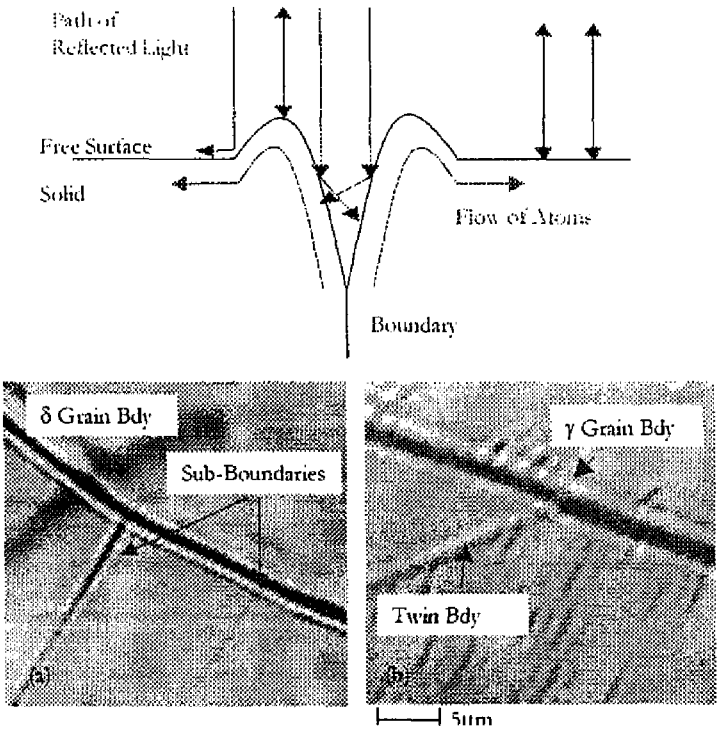


FIGURE 2 Schematic representations and examples of interfaces observed with LSCM

A finite time is required for thermal etching to form a groove and therefore rapidly moving boundaries are difficult to detect. The appearance in the laser scanning confocal microscope of a moving grain boundary is very similar to that of a moving interphase boundary, and it is therefore very important to be able to distinguish between grain growth and interphase boundary movement. Consequently, if sufficient time is allowed for grain growth to cease before a phase transformation occurs, this ambiguity is eliminated and the course of events can be interpreted with confidence.

Sub-grain structures in delta-ferrite

Typical microstructures of the plain carbon steels shown in Table 1. following heating into the delta-ferrite single phase region, are presented in Figure 3. A fine network of sub-boundaries within delta-ferrite grains is observed. Delta-ferrite grain boundaries are high angle interfaces and develop a pronounced thermal groove, dark black lines, and form triple points of 120° with each other. Sub-boundaries on the other hand, are low energy boundaries, therefore do not develop a substantial thermal groove, and appear as faint lines contained within the delta-ferrite grains. Additionally, where they intersect the heavily grooved delta-ferrite grain boundaries, 120° triple points are not formed due to the lower energy of the sub-boundaries.

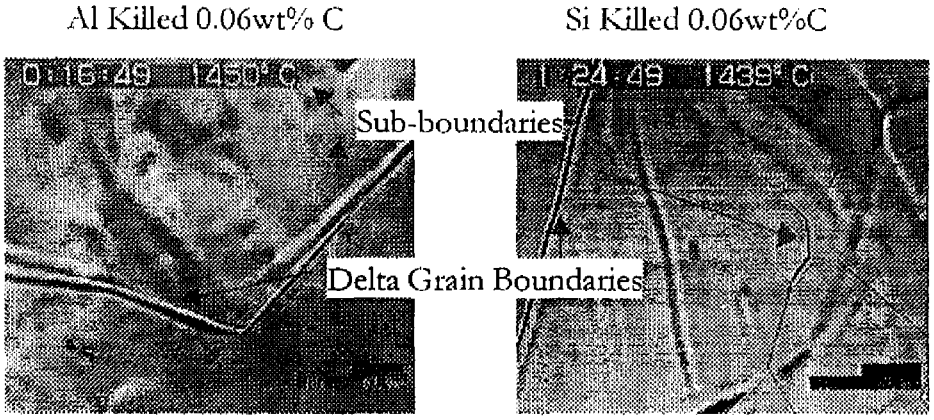


FIGURE 3 Sub-grain structures in Al and Si Killed 0.06wt%C Steels

In both the images presented in Figure 3, the surface appears quite rough. This roughness is primarily a result of the presence of prior grain boundaries, either austenite grain boundaries that existed before the transformation, or delta-ferrite grain boundaries before grain growth occurred. This is particularly evident in the silicon-killed specimen where a wide dark line runs through the middle of the delta-ferrite grain. This feature can be distinguished as a prior grain boundary by the lack of definition, compared to delta-ferrite grain or sub-boundaries. The action of surface diffusion is to flatten the thermal groove associated with prior grain boundaries. Once the sub-structure has developed, it appears to be quite stable. In Figure 4, the microstructure was recorded over a period of two minutes and during this time very little change can be detected in the sub-boundary microstructure.

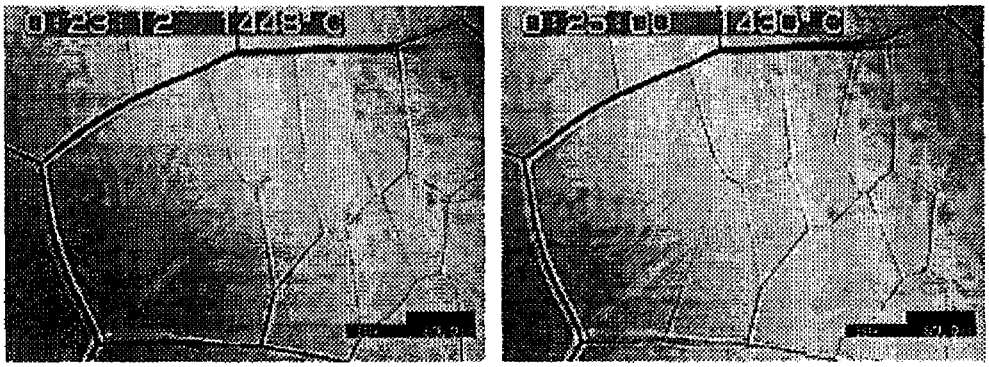


FIGURE 4 Sub-grain structure in a 0.06%C Si-killed steel, observed in the LSCM

Estimation of sub-boundary energy

In an attempt to estimate the sub-grain boundary energy, force balance calculations were conducted on the triple points of delta-ferrite grain boundaries and sub-boundaries. The triple points selected for measurement were restricted to those sufficiently spaced from adjacent triple points to eliminate interactions that could effect the equilibrium shape. The delta-ferrite grain boundary energy was taken as $0.471 J/m^2$, Yin et al[13]. The sub-boundary energy is calculated by the energy balance expressed in Equation 1 and the values have been recorded in Table 3. Although there is considerable variation in the calculated sub-boundary energies, the sub-boundary energy is substantially lower than the grain boundary energy, varying between 7% and 36% that of a delta-ferrite grain boundary.

$$\sigma_{SB} = 2\sigma_{GB}\cos\left(\frac{\theta}{2}\right)$$ Equation 1

TABLE 3 Calculated range of sub-boundary energies

Steel	Minimum J/m^2	Maximum J/m^2	Number of Measurements
Al Killed	0.051	0.17	15
Si Killed	0.035	0.145	27

Sub-boundary formation in type 3CR12 stainless steel

The initial transformation from delta-ferrite to austenite on cooling a type 3CR12 stainless steel into the two phase delta-plus-gamma field at 1100°C occurred predominantly along the delta-ferrite grain boundaries, with a rim of austenite covering the boundaries. However, in some instances plates of austenite grew into the matrix of the delta-ferrite grains. Figure 5 (a) shows austenite formation on delta-ferrite grain boundaries and also austenite plates that have grown into the delta matrix. On reheating to 1400°C, within the delta-ferrite single-phase region, the austenite reverts back to delta-ferrite, as shown in Figure 5 (b). A network of sub-boundaries forms only in positions where plates of austenite were present at 1100°C. The tip of the austenite plate is expected to be associated with a stress concentration and multiple sub-boundaries are running off from the tip of the plate into the matrix of the delta-ferrite grain. These observations provide convincing experimental evidence, albeit indirect, that the transformation from austenite to delta-ferrite generates a sufficiently complex dislocation structure to enable recovery to occur, and through the process of polygonisation leads to the formation of low angle boundaries (sub-boundaries).

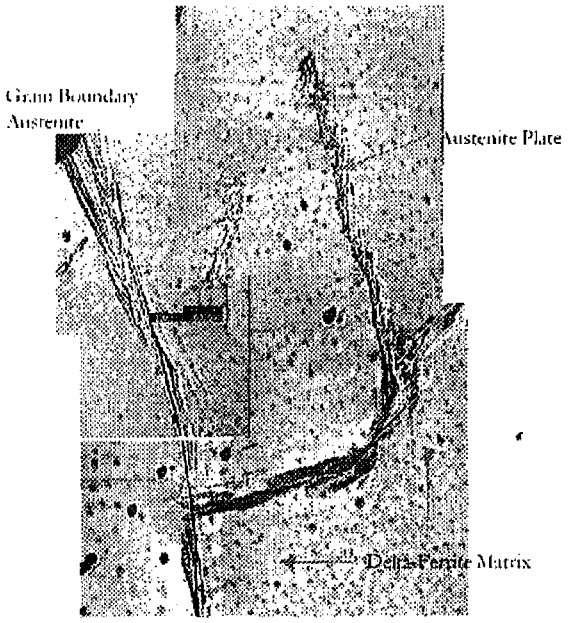


FIGURE 5 (a) Austenite plate growth into delta-ferrite matrix (LSCM, 1100°C). 3CR12 steel on cooling

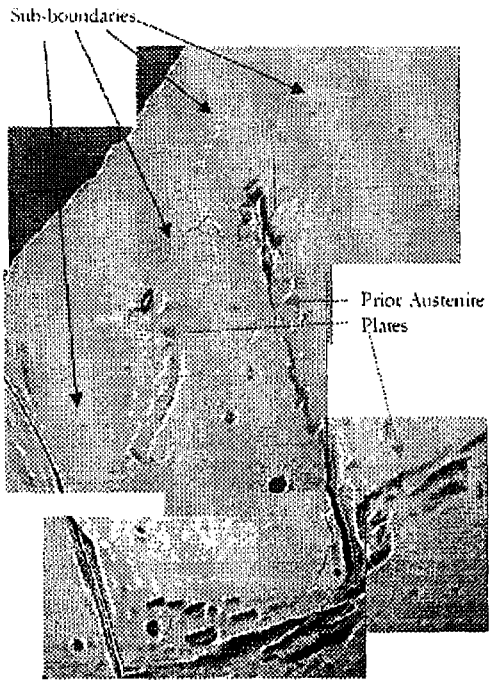
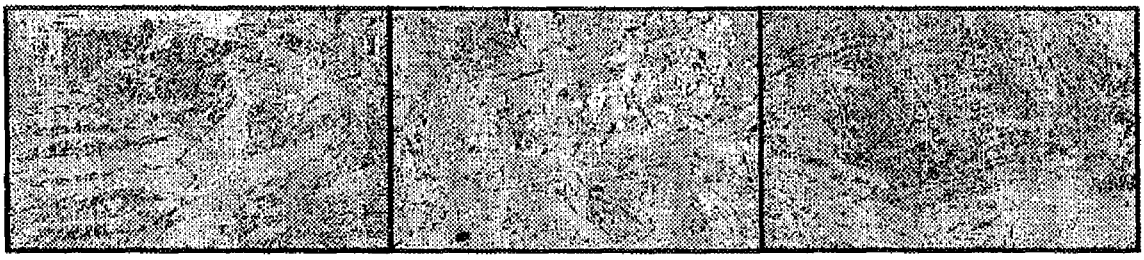


FIGURE 5 (b) Localised recovery structure (LSCM, 1400°C). 3CR12 on heating

Influence of thermal cycling through the delta/gamma phase transition on austenite decomposition in low carbon steel

Thermal cycling through the delta-to-gamma phase transition leads to surface roughening to the extent that the image becomes diffuse and hence, clear LSCM micrographs could not be obtained in the thermal cycling experiments. Therefore optical metallography was used to characterise the microstructure following austenite decomposition, although the thermal cycling was done in the laser microscope. Following the thermal cycles shown in Table 2, samples were mounted in Bakelite, polished, etched in nital and the microstructure assessed by optical microscopy. The change brought about in the microstructural development on austenite decomposition by the thermal cycling experiments outlined above are summarised in Figure 6. In this figure the microstructures of specimens form from Tests 1, 3 and 5 are shown. This experimental evidence clearly shows that the final microstructure following decomposition of austenite below 900°C, is influenced by thermal cycling through the delta/gamma phase transition occurring at a temperature of approximately 1400°C.



(a) Test 1 (0 sec hold) (b) Test 2 (20 sec hold) (c) Test 3 (60 sec hold)

FIGURE 6 Microstructural response to thermal cycling, 0.06%C steel

The microstructure following zero-soak time cycle, Test 1, is characterised by large, blocky structures. Widmanstätten and bainitic ferrite predominate over allotriomorph ferrite, frame (a). No pearlite is present, excess carbon has been rejected as inter and intra plate carbides. The microstructure obtained by soaking times of 20 seconds at 1350°C and 1450°C respectively is presented in frame (b), Test 3. The structure is comprised of polygonal ferrite and pearlite with a greatly reduced proportion of Widmanstätten ferrite and bainite. The microstructure is distinctly different from that shown in frame (a). Considering that the only variable changed was the soak time at 1350°C and 1450°C, the formation of the microstructure observed in Figure 6 (b) cannot be explained unless the austenite grain size of Test 3 is smaller than that of Test 1. This observation provides evidence in support of the premise that the development of a delta-ferrite recovery structure leads to the refinement of the austenite grain size when delta-ferrite transforms to austenite. The microstructure obtained by soak times of 60 seconds at 1350°C and 1450°C, frame (c), is characterised by large colonies of bainite and Widmanstätten ferrite, a small amount of pearlite is present but inter- and intra-granular carbides dominate. Some polygonal ferrite grains are observed. This microstructure is again distinctly different from those shown in Figure 6 (a) and (b). The estimated volume fraction of Widmanstätten/bainite and polygonal ferrite/pearlite components in each microstructure are shown in Table 4. The Vickers hardness of the five samples was measured with a load of 10kg. The hardness and standard deviations are tabulated in Table 4. Despite the large standard deviations in the measured hardness numbers, the average measured hardness is statistically different. This analysis provides a quantifiable measure to indicate that not only is the thermal treatment modifying the microstructure but also the mechanical properties.

TABLE 4. Volume percentage and Hardness of austenite decomposition products for the silicon-killed steel shown in Table 1

Soak time	Widmanstätten/ Bainite	Polygonal Ferrite / Pearlite	Hardness	
seconds	volume %	volume %	Ave. VHN 10	Standard Deviation
0	99	1	123.4	3.497
10	92	8	121.4	3.851
20	6	94	114.0	2.732
30	34	64	118.4	4.177
60	95	5	120.1	3.341

DISCUSSION

Although laser-scanning confocal microscopy seems eminently capable of being used as a technique to study events occurring at high temperature, care has to be taken in the interpretation of observations made. Phelan[14] has recently shown that there exists a nexus between phase transformations observed on the free surface in LSCM and events occurring in the bulk. On the other hand, grain boundary movement and precipitation of non-metallic inclusions are influenced by the nature of the free surface and observations in the LSCM do not necessarily correlate with bulk behaviour. A particularly useful feature of LSCM is the formation of thermal grooves at line defects. These grooves delineate pre-existing features and can be usefully employed, for example, by studying subsequent phase-boundary movement.

Low energy sub-grain boundaries have been observed in the delta-ferrite phase of aluminium as well as silicon-killed low carbon steels. These sub-grain boundaries, once formed are quite stable but are easier to observe in silicon-killed steel than in aluminium-killed steel. They are also much more distinct in the silicon-killed steel. This observation is specifically interesting because Yin et al [13], who studied the delta-to-gamma transformation in aluminium-killed steels by in-situ observation, did not report the presence of such delta-ferrite sub-boundaries. It is possible that they may not have observed these sub-boundaries because the sub-boundaries are very faint in aluminium-killed steel. We have proposed that these sub-grains form in the delta-ferrite matrix by the re-arrangement of dislocation networks by a process of polygonisation. It was further proposed that these dislocations are generated by a combination of transformation stresses and the difference in thermal expansion coefficients of the delta and gamma phases.

It is clearly important to seek experimental evidence in support of the premise that transformation stresses induce dislocation generation which, in turn, result in polygonisation. It is specifically important to prove that such a dislocation structure can form on heating austenite into the delta-ferrite phase field. It is not possible to observe dislocations in the laser-scanning confocal microscope and transmission electron microscopy at the high pertaining temperature is not possible either. However, Phelan[14] has shown that thin austenite plates form on partially transforming delta-ferrite in a type 3CR12 steel. It is expected that a significant stress concentration would be present at the tip of such a plate and it is thought that this stress concentration, combined with the transformation stresses may generate dislocation networks on re-heating into the delta-phase field. Should such dislocation networks form polygonisation may occur. This seems to be what has happened in the specimen shown in Figure 5. Sub-grain boundaries have formed in the area around the tip of the plate where the highest stress concentration is expected. Although this observation was made in a ferritic stainless steel, it seems reasonable to expect that dislocation networks may also form during the transformation of austenite to delta ferrite in plain carbon steel.

We have also proposed that the final microstructure in low carbon steel, resulting from the decomposition of austenite, may be influenced by the existence of a sub-grain structure in the delta phase. Convincing experimental evidence was provided in support of the premise that repeated cycling through the delta/gamma phase transition would develop a refined sub-grain structure in the delta-phase. This refined delta sub-grain structure will lead to refinement of the gamma structure, which in turn, will lead to a modified ferrite structure on decomposition of the austenite. This is an exciting prospect because it means that the formation of a recovery structure in delta-ferrite could in principle provide a means of microstructural modification and control of the final product in the absence of mechanical deformation. Such a procedure would have specific relevance to the strip casting of low-carbon steels where control of microstructural development through thermo-mechanical processing is not possible. Additionally, because the strip-cast product is thin steel sheet it can be heated and cooled very rapidly, and hence, thermal cycling can be done more easily than with conventionally continuously cast steel.

The development of sub-boundaries through recovery does not occur instantaneously and sub-boundaries develop only as a function of time. During this time the delta-ferrite grains may grow by the normal mechanism of grain growth and hence there are two competing processes at play: sub-grain formation and delta-ferrite grain growth. A well-developed sub-grain structure will refine the ensuing austenite grain size while delta-ferrite grain growth will have the opposite effect. The kinetics of neither process has been adequately studied in delta-ferrite and hence, only qualitative conclusions can be drawn. However, it is reasonable to assume that these two competing processes will result in an initial refinement of the microstructure as sub-boundaries develop, but with time, delta-ferrite grain growth would dominate, reducing the number of nucleation sites available for austenite formation, not only through a reduction in the grain boundary area, but also through sweeping up the sub-boundaries themselves. This proposed sequence of events can explain the initial refinement and coarsening at longer soak times, of the microstructures shown in Figure 6. In the absence of quantified kinetic data on sub-grain formation and delta-ferrite grain growth, the proposed sequence of events provides a plausible explanation for the observed microstructural changes.

CONCLUSIONS

- Sub-boundaries have been observed in the delta-ferrite phase of low carbon steels.
- It is proposed that dislocations are generated by the strains associated with the austenite to delta-ferrite transformation on heating. These dislocations eventually form sub-boundaries through the process of polygonisation.
- Experiments conducted with a ferritic stainless steel provided evidence in support of this proposed mechanism.
- Experimental evidence was found for the proposition that the sub-boundary structure in delta-ferrite can play a role in modifying austenite decomposition products.
- The ability to control microstructural development without recourse to thermo-mechanical processing could in principle provide a novel means of microstructural modification and control, especially in near-net shape casting processing.

REFERENCES

1 H.I. Aaronson, "Nucleation kinetics of grain boundary ferrite allotriomorphs at austenite grain faces during the proeutectoid ferrite reaction in Fe-C, Fe-C-X and Fe-C-Mn-X alloys", Materials Forum (1999), vol. 23, pp. 1-22

2 P.J. Clemm and J.C. Fisher, "The influence of grain boundaries on the nucleation of secondary phases", Acta Metallurgica, Vol. 3, Jan., (1955), pp. 70-73

3 W. Huang and M. Hillert, "The Role of Grain Corners in Nucleation", Metallurgical and Materials Transactions A, Vol 27A, Feb., (1996), pp. 480-483

4 M. Enomoto, W.F. Lange and H.I. Aaronson, "The kinetics of ferrite nucleation at austenite grain edges in Fe-C and Fe-C-X alloys.", Metallurgical Transactions A, V17A, Aug (1986), pp. 1399-1407

5 M.Ferry, M. Thomson and P.A. Manohar, "Decomposition of coarse grained austenite during accelerated cooling of C-Mn steels", ISIJ, vol 42 (2002), pp86-93

6 K. Mukunthan, L. Strezov, R. Mahapatra and W. Blejde, "Evolution of Microstructures and Product Opportunities in Low Carbon Steel Strip Casting", Proc. Brimacombe Memorial Symposium, Ed. G.A. Irons and A.W. Cramb, Oct., 2000, Vancouver, Met-Soc, pp. 421-438

7 N.P. Goss, "Subboundary Structures of Recrystallised Iron", Transactions of AIME, Vol. 145, (1941), pp. 272-280

8 J.J. Hauser, J.M. Capenos and B.R. Banerjee, "Submicroscopic structures in tempering 410 stainless steel", ASM Transactions Quart., Vol. 54. (1961), pp. 514-525

9 A.J. Baker, P.M. Kelly and J. Nutting, "Structures Resulting from Phase Transformations in Steels", Electron Microscopy and Strength of Crystals, ed. G. Thomas and J. Washburn, University of California, Berkeley, July 5-8, 1961, pp. 899-916

10 J. Talbot, "The annealing behaviour of high purity iron", Recovery and Recrystallisation of Metals, Interscience Publishers, New York, N.Y., (1963), pp. 269-303

11 T. Furuhara and T. Maki, "Variant selection in heterogeneous nucleation on defects in diffusional phase transformation and precipitation", Materials Science and Engineering A, Volume 312, Issues 1-2, 15 August 2001, pp. 145-154

12 Zapsukalov and Dippenaar [2001]; Private communication.

13 H. Yin, T. Emi and H. Shibata, "Morphological Instability of δ-Ferrite / γ-Austenite interphase Boundary in Low Carbon Steels", Acta Materialia, Vol. 47, No. 5 (1999), pp. 1523-1535

14 D. Phelan, PhD thesis, University of Wollongong, 2002