Particle-stimulated Nucleation of Ferrite in Heavy Steel Sections

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In heavy steel sections, strength and toughness are improved remarkably by refining and homogenizing the final ferrite microstructure. The production of heavy sections by universal-type rolling mills takes place at elevated temperature followed by slow cooling rates. Therefore, refinement of the ferrite microstructure through modern thermomechanical, TMP, techniques is not feasible. This paper explores and presents the use of particle-stimulated nucleation of ferrite, the PSN (particle-stimulated nucleation) mechanism, to refine the ferrite grain size and eliminate the high-carbon, low transformation products otherwise found in transformed coarse-grained austenite. In this study, MnS and Ti-oxide particles were used to promote intragranular ferrite, or IGF, in a typical ASTM A572 grade 50 Steel. This work included the study of the decomposition behavior of coarse grained austenite as a function of very slow cooling rate. In addition, the nucleation of ferrite grains taking place at γ/γ, γ/α and γ/inclusion interfaces was identified and quantified.

KEY WORDS: grain refinement; intragranular ferrite (IGF); particle-stimulated nucleation (PSN); phase transformation.

1. Introduction

The principal goal of TMP in ferrite-pearlite steels is to achieve a predetermined austenite microstructure prior to transformation to insure the finest ferrite microstructure for a given cooling rate. The austenite grain refinement found in TMP is obtained from controlling the recrystallization and subsequent grain coarsening of austenite in a multipass rolling schedule.1–4) Tanaka1) showed that TMP of plates could be considered as generally consisting of the controlled rolling process followed by interrupted accelerated cooling (IAC). The major purpose of the controlled rolling process is to produce deformed austenite to increase the density of ferrite nucleation sites, while IAC enhances the ferrite nucleation rate.

In the case of heavy plates where the final thickness is larger than 40 mm, the effects of controlled rolling and accelerated cooling will not be spread equally over the whole thickness of the plate. This could give a large variation in the final microstructure and produce a coarse grain structure and deteriorating strength and toughness at mid-thickness of the plate section. The limitation of applying the traditional TMP in the case of heavy sections is based on three main reasons. One is the regional differences in the thickness which give a large variation in rolling temperature and reduction ratio among the different regions of the given section during the hot rolling process. The second reason is due to the limitation of the rolling equipment, i.e., motors, gears and stands, to give large reduction per pass for such heavy sections as required in the traditional TMP. The third source of limitation comes from the difficulty of applying accelerated cooling processes to these heavy sections. This is because of the difference between the cooling rate at both the center and the surface will lead to different transformation products and, consequently, different mechanical properties. Moreover, the transformation of coarse Dγ under rapid cooling rate could result in undesirable high carbon non-polygonal ferrite microstructures of high yield-to-tensile ratio and lower notch toughness.5–7)

The production of heavy sections by universal-type rolling mills, which are limited in rolling load and torque capacity, creates a special environment of high deformation temperature with small reduction per pass, large interpass time, large number of passes, and long rolling times. All of these processing parameters contribute to coarsening the equiaxed, recrystallized austenite grains prior to the transformation. Considering the final product thickness, Fig. 1 shows schematically the conditions under which fine ferrite grains can be obtained from a given austenite grain size. The two main parameters that affect the final ferrite grain size are the microstructural state of the austenite prior to the phase transformation and the cooling rate through the phase transformation.7)

Based on the above facts, it seems to be impossible to produce fine ferrite microstructures in heavy sections by using the conventional, well-established TMP approach which depends on effective austenite conditioning and interrupted accelerated cooling. Therefore, another approach is needed to refine the final ferrite structure. In the approach adopted here, the traditional $S_\gamma$ of the austenite is
augmented by an additional $S_v$ coming from ferrite nucleating particles located inside the original austenite grains. This has come to be known as particle-stimulated nucleation or PSN, which leads to the formation of intragranular ferrite, IGF. This approach has been studied by several workers\textsuperscript{7–18) who proposed using second phase particles to promote the nucleation of ferrite. The use of non-metallic inclusions as IGF nucleation sites was originally studied to refine $D_a$ from coarse $D_g$ in the heat affected zone, HAZ, of weldments.\textsuperscript{19–30) Adding active second phase particles to steel increases the $S_v$ value beyond that associated with austenite grain boundaries and provides additional nucleation sites to form fine ferrite grain size during transformation.

2. Role of Inclusions in the IGF Formation

Although the mechanism by which inclusions nucleate both acicular and polygonal IGF is yet unclear, there have been four mechanisms suggested for this nucleation\textsuperscript{16,18,31)}: 1) Simple heterogeneous nucleation at inclusions; 2) Lattice matching/epitaxial growth; 3) Nucleation assisted by volumetric strain; and 4) Nucleation assisted by local solute depletion effects.

However, the primary role of inclusions is to provide heterogeneous nucleation sites for ferrite formation beside those available at the austenite grain boundaries.\textsuperscript{11,32)} Several factors are known to influence the formation of IGF during the phase transformation. These factors include: number density of inclusions,\textsuperscript{20,22)} size,\textsuperscript{20,33,34)} and type of inclusions,\textsuperscript{21,23,24)} prior austenite grain size,\textsuperscript{8,20,29,30)} hardenability,\textsuperscript{19,20)} and cooling rate.\textsuperscript{20)} It is generally believed that increasing either the inclusion density per unit volume or prior austenite grain size tends to promote the formation of IGF.\textsuperscript{20,21)} The degree of inter- and intragranular nucleation may change by altering the prior austenite grain size, where small prior austenite grains may favor the grain boundary reaction, and the intragranular polygonal ferrite will dominate over intragranular ones.\textsuperscript{9,14,15)} According to Ricks \textit{et al.},\textsuperscript{21)} coarse inclusions are theoretically expected to be more effective nuclei. This prediction has been confirmed experimentally by several workers\textsuperscript{20,33,35)} Thewlis \textit{et al.},\textsuperscript{33)} showed that inclusions larger than 3 $\mu$m act as effective nucleation sites for IGF.

In this work, the MnS and Ti–oxide inclusions were used to promote intragranular ferrite (IGF) nucleation in a typical ASTM A572 grade 50 Steel. Both types of inclusions have reported as being highly effective sites for IGF nucleation.\textsuperscript{15,24,36–41)} This work included the study of the decomposition behavior of coarse grained austenite and nucleation of the IGF as a function of temperature at very slow rates of cooling. In addition, the effects of the inclusions type, size and volume fraction on the kinetics (nucleation and growth) of IGF were investigated.

3. Experimental Procedure

A typical ASTM A572 grade 50 steel (Jumbo steel) was selected to be the reference material in this investigation. The Jumbo steel samples were prepared from a commercial, continuous cast slab section. Based on the chemical composition of the Jumbo steel, two different steels were designed and laboratory melted to have abnormally high volume fractions of MnS and Ti–oxides inclusions. The chemical compositions of these three steels are listed in Table 1. The thermal processing cycle that was applied to these materials is schematically shown in Fig. 2. This thermal processing schedule attempts to simulate as closely as possible the thermal cycle of the hot rolled sections in the universal mill. It is important to note that the very slow rate of cooling used, 0.08°C/s, eliminated any effect of prior hot deformation. Hence, only heat treated samples were used in this part of the study.\textsuperscript{42)}

Samples of $12 \times 12 \times 6$ mm from the three steels were heated and held for 60 min at different temperatures. The reheating temperature for each steel was selected to be above its respective grain coarsening temperature. During the slow cooling procedure, samples were water quenched at 10°C intervals starting from 800°C and ending at 500°C. These temperatures were selected to reveal the nucleation and growth of the allotropic and idiomorphic ferrite. The quenched specimens were longitudinally cut in half and

![Fig. 1](image1.png)  
**Fig. 1.** The relation between the austenite grain size and cooling rates for achieving fine ferrite ($D_a = 15 \mu$m).

![Fig. 2](image2.png)  
**Fig. 2.** Schematic diagram of the thermal processing of the three steels.

| Table 1. Chemical composition of the Jumbo, MnS and TiO steels. |
|------------------|--------------------|-----------------|-----------------|-----------------|
| Steel            | C      | Si     | Mn      | P | S | Al | Cu | N | O  | Nb | Ti | V |
| Jumbo           | 0.08%  | 0.3%   | 1.42%   | 0.008 | 0.003 | 0.034 | 0.31 | 0.005 | - | 0.003 | 0.003 | 0.132 |
| TiO             | 0.095% | 0.03% | 1.22%   | 0.008 | 0.003 | 0.003 | 0.28 | 0.004 | 0.022 | - | 0.02 | 0.14 |
| MnS             | 0.086% | 0.32% | 1.57%   | 0.008 | 0.037 | 0.028 | 0.27 | 0.005 | 0.002 | - | - | 0.137 |

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metallographically prepared for further analysis. Furthermore, a sample of each type of steel was cooled down to room temperature to measure the final ferrite grain size and to evaluate the ultimate refining of the ferrite microstructure resulting from the PSN.

Quantitative measurements were performed using a computer controlled Bioquant IV image analysis system attached to an optical microscope. SEM and EDX spectroscopy were used for the characterization of the microstructures and for qualitative microanalysis. The analyses were conducted using a Philips XL 30 FEG scanning electron microscope (SEM) operated at 10–15 kV (accelerating voltage). This instrument was interfaced to an EDAX energy dispersive X-ray spectrometer and analysis system.

4. Results and Discussion

4.1. Inclusion Characterization

The steel samples were examined using optical and scanning electron microscopy to characterize the non-metallic inclusions. Although the volume fraction, size and distribution varied widely, the majority of the inclusions in the three steels have a nearly spherical shape, Fig. 3. The Ti–oxide inclusions show rough and porous surfaces, while the MnS inclusions in the Jumbo and MnS steels have smoother surfaces. The quantitative analysis of inclusions in these three steels, including the volume fraction, mean size and the number of particles per unit area is listed in Table 2.

In order to study the IGF nucleation behavior, the volume fraction of inclusions was increased from 0.012% in the Jumbo steel to 0.146% and 0.332% in the TiO and MnS steels, respectively. The mean size of inclusions in the Jumbo steel was 1.37 ± 1.08 μm and the size ranged from 0.18 to about 6.0 μm. In the TiO steel, the mean size of inclusions decreased to 1.08 ± 1.51 μm while the size range expanded between 0.12 and 21.0 μm. In the MnS steel, the mean size of inclusions was 1.33 ± 0.97 μm while the size range extended between 0.16 and 10.0 μm. In both the Jumbo and MnS steels, approximately 50% of the inclusions were smaller than 1.0 μm, while the majority, 75%, of the inclusions in the TiO steel was smaller than 1.0 μm. The amount of coarse inclusions that are larger than 6.0 μm was limited in all three materials. It only corresponds to 1.3% and 0.15% of the total observed inclusion sizes in both TiO and MnS steels, respectively. All observed inclusions in the Jumbo steel are smaller than 6.0 μm. The overall size distribution of the three steels for inclusions up to 6.0 μm is shown in Fig. 4. Regardless of the volume fraction of the inclusions, the MnS particles in both Jumbo and MnS steels have similar mean size and size distribution.

Two main types of inclusions were found in the TiO steel. The first type consists of inclusions smaller than 3.0 μm in size, while the other type consists of inclusions larger than 3.0 μm, Table 2. Figure 4 shows the size distribution of the three steels with inclusions up to 6.0 μm.

Table 2. Quantitative analysis of inclusions in the three steels.

<table>
<thead>
<tr>
<th>Inclusions</th>
<th>Materials</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Jumbo</td>
</tr>
<tr>
<td>Mean size (μm)</td>
<td>1.37 ± 1.08</td>
</tr>
<tr>
<td>Inclusions &lt; 1.0μm</td>
<td>46 %</td>
</tr>
<tr>
<td>Volume fraction (Vd)</td>
<td>1.2x10⁻⁵</td>
</tr>
<tr>
<td>Number density per unit area (N0, μm⁻³)</td>
<td>5.1x10⁴</td>
</tr>
<tr>
<td>Number density per unit volume (N0, μm⁻³)</td>
<td>3.72x10⁴</td>
</tr>
</tbody>
</table>

Fig. 3. SEM micrographs showing the morphology of inclusions in the three materials.

Fig. 4. Size distribution of the three steels with inclusions ≤6.0 μm.
larger than 3.0 μm. The EDX spectrum in Fig. 5 shows that the majority of the small inclusions present in this steel are mainly Ti–oxide with small amounts of Mn. In addition to the Ti–oxide, the EDX spectra in Fig. 6 show that the large inclusions usually contain complex compounds such as Mn, MnS and Al–oxides. The amount of these complex compounds varied widely from one particle to the other and even within the same particle. On the other hand, the EDX analysis of the particles in the Jumbo and MnS steels shows that the majority of inclusions presented in these steels are mainly MnS.

4.2. Thermal Processing Analysis

In the beginning of the thermal processing study, samples were held for 60 min at 1250, 1300 and 1350°C for the Jumbo, TiO and MnS steels, respectively. These temperatures were selected to be above the grain coarsening temperature of each material. The austenite grain size prior to the phase transformation was measured for each material to determine the relation between the decomposed austenite and nucleated ferrite grain sizes. The average austenite grain sizes just before the phase transformation, at 825°C, are 372±160, 240±106 and 230±87 mm for the Jumbo, TiO and MnS steels, respectively. However, the measured value of the transformation temperatures for the Jumbo, TiO and MnS steels was 805, 805 and 810°C, respectively.

A series of optical micrographs depicting the decomposition behavior of the prior austenite microstructure and the formation of the ferrite microstructure as a function of the cooling temperature for the Jumbo, TiO and MnS steels are shown in Figs. 7–9, respectively. Although the optical micrographs show that the phase transformation of all steels started just above 800°C, each material behaved differently during the transformation.

In the Jumbo steel, Fig. 7, the sample quenched at 770°C shows the consumption of all available austenite grain boundaries with nucleated proeutectoid ferrite. There was no evidence of IGF formation at this temperature. Upon quenching from 750°C, additional nucleation and growth of ferrite along and perpendicular to the austenite grain boundaries was observed. Simultaneously, the IGF started to take place as a second layer of ferrite that nucleated at the previously formed γ/α interface. The growth of the primary nucleated ferrite grains at austenite grain boundaries continued and more IGF formed as the temperature decreased. Due to the low inclusion content, the majority of the observed IGF in the Jumbo steel was nucleated at the γ/α interface, while few nucleation events were observed within the austenite grains. The transformation appeared to be completed around 600°C, where the volume fraction and grain size of ferrite did not change considerably with subsequent reduction in the temperature. The formation of polygonal ferrite grains rapidly increased the carbon concentration within the center of the austenite grain, thereby reduc-
ing the undercooling making the subsequent nucleation events more difficult. At some certain point below the eutectoid temperature, the nucleation of ferrite grains stopped and the remaining untransformed austenite formed other low temperature microconstituents, as shown clearly in the optical micrographs of 600 and 500°C.

The prior austenite microstructure in both TiO and MnS steels decomposed in a very similar manner. In the TiO steel, Fig. 8, the consumption of the available prior austenite grain boundaries is observed around 760°C. Further nucleation and growth of ferrite along and perpendicular to the austenite grain boundaries were observed with cooling to 750°C. At this temperature, in addition to nucleation of ferrite at the γ/α interface, ferrite started to nucleate intragranularly at inclusions inside the austenite grains. On the other hand, the utilization of the available austenite grain boundaries was completed around 750°C in the MnS steel, Fig. 9. The further nucleation and growth of ferrite along the prior austenite grain boundaries were observed with cooling to 730°C. The differences between the MnS and TiO steels transformation temperatures could be explained due to the high content of Mn and Ti in these materials, respectively. Since solute Mn is considered as an austenite stabilizer, therefore, a higher degree of undercooling was required in the MnS steel to achieve a transformation effect similar to that found in the TiO steel.43)

With continuous cooling below 710°C, inclusions in both steels acted as effective nucleation sites for IGF, while the growth of the primary nucleated ferrite grains at austenite grain boundaries continued. Due to the high inclusion content, the volume fraction of ferrite grains increased remarkably between 710 and 690°C in both cases. The higher activity of the grain boundaries nucleation sites is due to the combination of (i) its lower free energy barrier to ferrite nucleation than intragranular inclusions,33) and (ii) its higher diffusivity. However, the transformation appeared to be completed at approximately 670°C, where the volume fraction and grain size of ferrite did not change much with subsequent reduction in the temperature. The low temperature microconstituents were not observed at low temperatures due to the relatively small austenite grain size and high inclusion volume fraction in both materials.

Graphs of the variations of the total ferrite volume fraction as a function of quenching temperatures in the three steels are depicted in Fig. 10. The comparison between the three curves exhibited the similarity in the decomposition behavior of the TiO and MnS steels. The obvious differences between the decomposition curve of the Jumbo steel
and the curves of the PSN materials reflect the significant effect of the inclusions on the kinetics of the decomposition behavior of austenite in the three steels.

4.3. IGF Nucleation Behavior

The study of the decomposition behavior of the austenite during the controlled cooling process confirmed that the nucleation of IGF was significantly stimulated below 720°C for TiO and MnS steels. However, the insightful investigation of the IGF nucleation behavior showed that the nucleation at the γ/I interfaces was started at higher temperatures. As shown in the SEM micrographs of Fig. 11, the first IGF nucleation event in the TiO and MnS steels was noticed at 750 and 740°C, respectively. At these temperatures, the nucleation of IGF was noticed at the large inclusions that ranged above 10.0 μm in the TiO steels and above 4.0 μm in the MnS steel. Due to the small surface curvature, the large inclusions are usually more effective in heterogeneous nucleation than the smaller ones.

It was noticed that the large Ti–oxide inclusions (>3.0 μm) was usually associated with nucleation of more than one ferrite grain, while the small inclusions commonly nucleated one ferrite grain, Fig. 12. This could be a result of the large surface area of the large inclusions which provide sufficient nucleation sites for more than one ferrite grain. On the other hand, MnS inclusions were occasionally asso-

Fig. 9. Decomposition behavior of the prior austenite microstructure in the MnS steel as a function of cooling temperature.

Fig. 10. The variation of total ferrite volume fraction as a function of cooling temperature in the three steels. Samples were cooled from 1250, 1300 and 1350 to 825°C at 0.24°C/s and then at 0.08°C/s for the Jumbo, TiO and MnS steels, respectively.

Fig. 11. IGF nucleation at the (γ/I) interface in Ti–oxide inclusions at 750°C in the TiO steel and in MnS inclusion at 740°C in the MnS steel.

Fig. 12. Nucleation of IGF at the Ti–oxide inclusions in the TiO steel. Small inclusion (2.5 μm) nucleated one ferrite grain at 730°C, while large inclusion (8.3 μm) associated with more than three ferrite grains at 740°C.
The effectiveness of the Ti–oxide particles could be attributed to the following five factors:

- The availability of significantly large inclusions in the TiO steel. The measurement of particles size shows that approximately 1.3% of Ti–oxide particles are above 6.0 μm, while only 0.15% of the MnS inclusions are above this range. The effect of the inclusion size has been confirmed by several workers.20,21,31,35,44)
- The hardenability effect.19,43) Ti–oxide particles promote the nucleation of ferrite, since solute Ti is classified as a ferrite stabilizer.
- Availability of the complex compounds in the Ti–oxide inclusions such as aluminum oxides, since solute Al is also considered as a ferrite former.23)
- Multi-nucleation events at large Ti–oxide particles, where one inclusion could nucleate several ferrite grains.23,25) This type of nucleation was rarely observed in the MnS steel.
- The rough and porous surface of the Ti–oxide inclusions could enhance the nucleation of IGF through decreasing the wetting angle in a similar manner to the heterogeneous nucleation in mould-wall cracks.45)

5. Conclusions

From the present study, the following conclusions can be drawn:

(1) The conventional TMP could not be applied as an austenite conditioning mechanism in the heavy steel sections. The refining of the ferrite microstructure coming from coarse-grained austenite can be achieved, through enhancing the IGF nucleation using the PSN mechanism.

(2) Ferrite nucleation and growth starts at prior γ/γ grain boundaries at high temperatures. With increased undercooling, the nucleation of ferrite at γ/α interfaces and at large intragranular inclusions occurs simultaneously. With falling temperatures, smaller inclusions are activated.

(3) The efficiency of the inclusion as a ferrite nucleation site is mainly related to the inclusion size and type. It was found that fewer than 14 and 50% of the total number of inclusion per unit area was contributing to the IGF nucleation in the TiO and MnS steels, respectively.

(4) The refinement of ferrite microstructure with the addition of inclusions could be due to the effect of the S_{C} value through: 1) Decreasing the D_{p} as a result of the pinning force effect, and 2) Increasing the IGF nucleation sites where the particles act as a heterogeneous nucleation sites.

(5) The high activity of Ti–oxide particles in nucleating IGF could be attributed to their: a) large size, b) complex compounds, c) hardenability, d) multi-nucleation characteristics, and e) surface morphology.

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Fig. 13. Nucleation IGF at a cluster of MnS particles in the MnS steel at 720°C.