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Grain growth in 18Ni 1800 MPa maraging steel

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The grain-growth behaviour in 18Ni8Co5Mo0.4Ti maraging steel was investigated in the temperature range 1123–1323 K. Grain sizes were estimated by measuring the diameter of the equivalent area of the individual grains directly on the optical microscope using a calibrated digital eyepiece. Grain-boundary migrations and substructure analyses were done using an electron microscope. These studies indicate that the overall grain growth in the steel follows the relationship $\Delta D = kt^n$ where ΔD is the increase in the grain size. However, during the initial stage, a "time lag" for the grain growth to start is observed which is attributed to the presence of highly dislocated austenitic matrix on annealing. Evidence of abnormal grain growth is also seen after annealing at 1173 K for 480 min and at 1123 K for 300 min. The growth exponents for the normal and abnormal grain growth were found to be 0.40–0.44 and 0.90–2.0, respectively. One significant deviation observed in the study was n decreasing from 0.44–0.40 at higher temperatures during the normal growth. This has been critically discussed in the light of the unique transformation characteristics of the steel. The activation energy for growth was calculated to be 60.0–62.5 kcal mol^{-1}, indicating the overall growth is controlled by self diffusion in γ-iron.

1. Introduction

Grain boundaries in polycrystalline materials act as barriers to crystallographic slip or shear and influence their phase transformation characteristics in several ways [1–5]. Grain-size control and its correlation with the mechanical properties has therefore been the subject of extensive studies. As most of the engineering alloys are processed in the temperature range where grain growth becomes imminent, understanding of the grain-growth behaviour constitutes an important area for the investigations. A number of growth models [6–10] have been developed over the last few years by considering energetics. Most of these are, however, confined to very simple systems such as those of high-purity metals, dilute solutions and/or precipitates in pure metals, and their applicability for the multicomponent systems is thus limited.

Maraging steel, being a high alloy steel and having unique transformation characteristics, may show quite complex growth behaviour because (a) the number of alloying elements present (totalling up to 30%) may interact differently with the migrating grain boundaries, (b) the grain-boundary precipitation of TiC/Ti(CN) is well established in the steel [11, 12] and would considerably influence the growth mechanism [13, 14], and (c) the highly dislocated austenitic matrix resulting from α–γ shear transformation on rapid heating to the annealing temperature [12] may change the nature of the driving force for growth [15–17]. Study of grain-growth behaviour in the steel, therefore, assumes a great theoretical interest, in addition to its being highly relevant for engineering processing, such as deformation through high-temperature routes, welding, etc. This paper presents a detailed study undertaken to establish the grain-growth mechanism in the steel under isothermal conditions.

2. Experimental procedure

All the investigations were carried out on samples taken from a double-vacuum melted 18Ni 1800 MPa maraging steel sheet of 2000 mm × 1000 mm × 1.2 mm. The chemical composition of the steel is given in Table I. Pieces 200 mm × 100 mm in size were cut from the sheet and solution annealed at 1093 K for 60 min to obtain fully recrystallized grains. Further analyses were made on flat square specimens of size 20 mm × 20 mm from the above solution-annealed samples.

Isothermal annealing was carried out in the temperature range 1173–1323 K at intervals of 50 K for the periods ranging from 15–600 min. All the thermal treatments were performed in a resistance-heated tubular furnace under a purified Ar atmosphere. Samples were introduced at the pre-fixed temperature and a minimum of 75 K s^{-1} heating rate was ensured to effect the α–γ transformation through a diffusionless process so that no compositional changes could occur even in localized areas. The mean grain diameter after each anneal was estimated by measuring the diameter of the equivalent areas of the individual grains directly on the optical microscope using a calibrated digital eyepiece. The number of observations was confined to 100 fields of view.

Microstructural examinations were carried out using optical and transmission electron microscopes. Optical metallographic samples were prepared by adopting standard mounting and polishing techniques

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TABLE I Chemical composition of maraging steel

Alloying elements (wt%)									Gases (p.p.m.)		
C	S, P (each)	Si, Mn (each)	Ni	Co	Mo	Ti	Al	Fe	N_2	O_2	H_2
0.004	0.005	0.001	17.7	7.8	4.4	0.45	0.1	Bal	30	20	2

and observations were made after etching with an aqueous solution of 10% ammonium persulphate. The grain boundaries were delineated by using 10% Nital as the final etchant. Thin foils for transmission electron microscopy (TEM) were prepared by the window technique using 10% perchloric acid and 90% methanol as the electrolyte. The polishing was carried out at 30 V, 233 K. The electron microscopic observations were done at 100 kV.

3. Results and discussion

The electron micrograph (Fig. 1) in the initial condition (solution annealed at 1093 K and air cooled) shows highly dislocated martensite and the mean diameter of the grains, as revealed by optical microscopy (Fig. 2), is of the order of 10 μm. It is also evident from Fig. 2 that the structure, apart from having hexagonal grains, consists of mixed grains with less than six sides (denoted *r*) and adjacent grains with more than six sides (denoted *p*). Energetically, for the arrays to be in equilibrium, the grains should be perfect hexagons in planar conditions. However, in practice this condition is rarely achieved and imperfect grains, becoming like *r* and *p*, are frequently observed. These imperfect grains disturb the triple point equilibria and grains become thermodynamically unstable. Grains such as *r* have high interfacial energy and their boundaries curve out in convex shapes to balance the energy. On annealing, these long boundaries try to straighten themselves to attain lower energy and in the process become consumed by grains such as *p*. If the required driving force is given, the growth process of larger grains at the expense of the smaller ones continues until they are inhibited by some opposing forces.

Figure 1 Transmission electron micrograph showing highly dense dislocation clouds in solution-treated condition (1093 K/1 h).

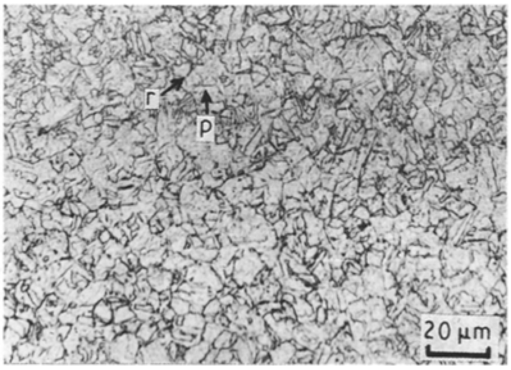

Figure 2 Initial microstructure showing grains with less than six sides (r) and more than six sides (p).

3.1. Growth behaviour

The progress of grain growth as a function of time during isothermal annealing is presented in Figs 3 and 4. It is seen that the growth rate during the initial stage of annealing at lower temperature is slow, whereas at 1273 K and above, it is quite rapid during the initial 60 min and continues monotonically thereafter (Fig. 3). In the classical grain growth model proposed by Beck *et al.* [7] the rate of grain growth is represented by the equation

$$D - D_0 = \Delta D = kt^n \quad (1)$$

where D is the average grain diameter at time, t, D_0 is the initial grain diameter and k and n are constants. To study the grain-growth behaviour in the steel, change in grain size, ΔD, was plotted against time on a logarithemic scale (Fig. 5). The plot exhibited linearity over a wide range of temperature and time, except for the initial and final stages of annealing at 1173 and 1223 K. During the initial stage, a "time lag", in line with that reported by Beck *et al.* in aluminium alloys

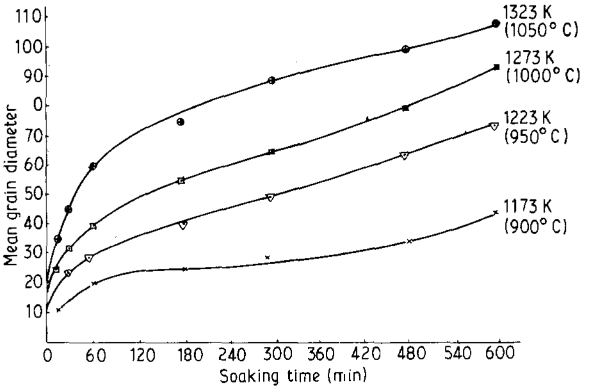

Figure 3 Isothermal behaviour of grain growth with time.

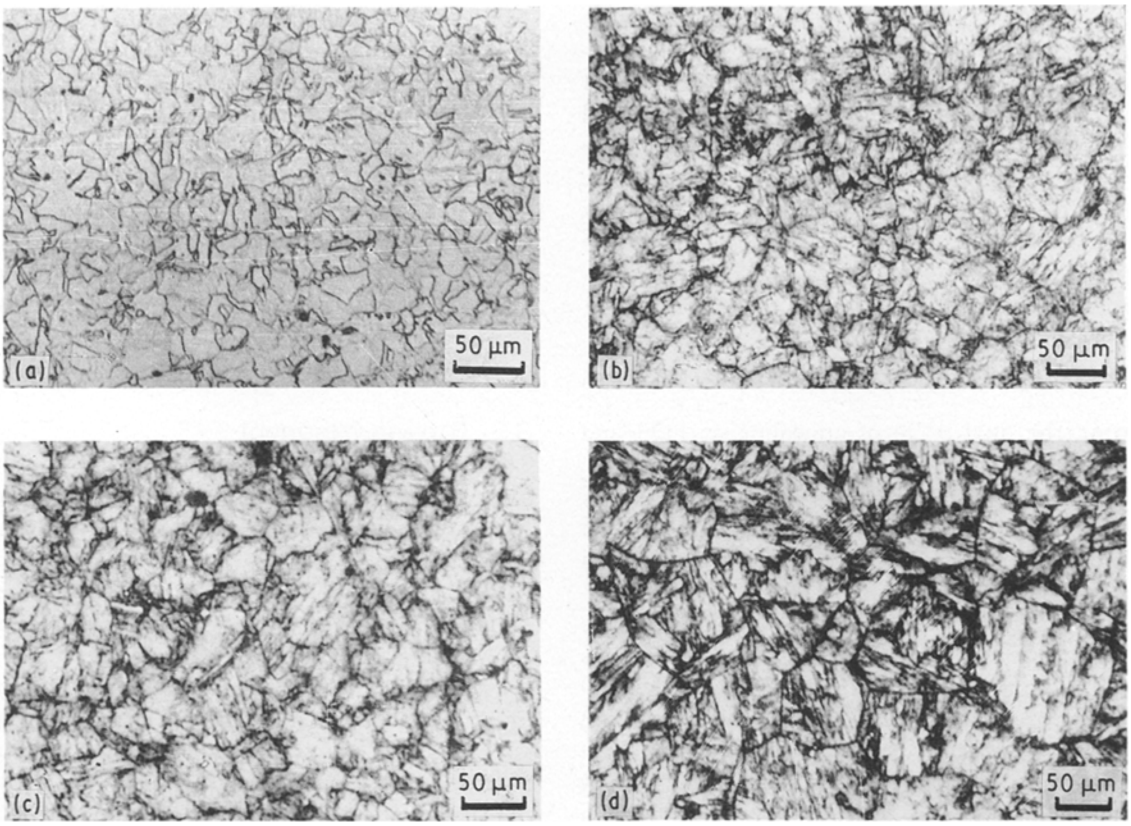

Figure 4 Typical microstructures showing progress of grain growth during isothermal annealing. (a) 1223 K/60 min, (b) 1223 K/480 min, (c) 1323 K/60 min, (d) 1323 K/480 min.

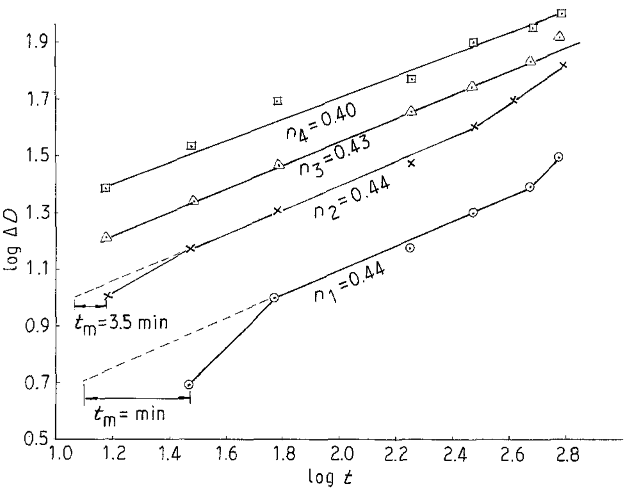

Figure 5 Log ΔD (increase in grain diameter) versus $\log t$ (soaking time, min) during isothermal grain growth at different temperatures. (○) 1173 K, (×) 1223 K, (△) 1273 K, (□) 1323 K.

[7], is observed. This deviation was attributed by these workers to the time of recrystallization of some deformed grains. However in the present case, as the initial samples were solution treated at 1093 K for 60 min, a "time lag" of 18 min at 1173 K cannot be totally accounted for by the recrystallization of the deformed grains. The observed longer time, nevertheless, can be explained on the basis of the unique transformation characteristics of the steel. It is well established [12] that in maraging steel highly dislocated austenitic grains are generated during rapid heating to the annealing temperature due to the α–γ shear transformation. The energy associated with these dislocations leads to the formation of subgrain boundaries on annealing through the process of polygonization. The electron micrograph in Fig. 6 confirms the presence of subgrain boundaries inherited from the parent austenite. Thus the total annealing time, t, during the initial stage must account for the time required for the development of substructure and can be written as

$$t = t_m + t_g \qquad (2)$$

where t_m and t_g represent the time for the formation of the substructure and actual growth, respectively. In the equation, t_m is the "time lag" referred to earlier,

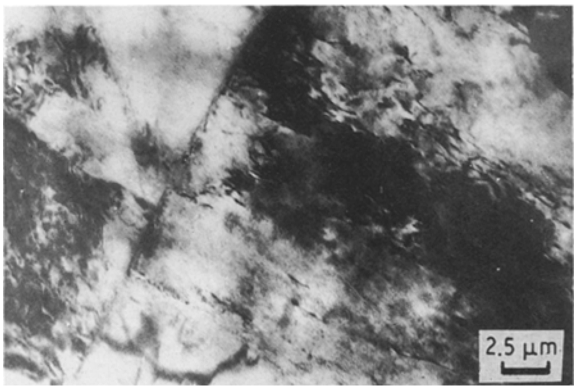

Figure 6 Transmission electron micrograph showing development of sub-boundaries after isothermal holding at 1173 K for 60 min.

and can be widely defined as the time required to annul the matrix inhibitions for the growth to set in. The definition is generalized considering the fact that in many systems, a number of other matrix-dominated factors such as recrystallization [7], deformation textures [17], etc., are also responsible for the delay in the actual growth. As evinced from Fig. 5, t_m sharply decreases with increasing annealing temperature (18 min at 1173 K and 3.5 min at 1223 K) and a near straight line relationship is obeyed as per Equation 1 at 1223 K and above, when

$$t_g \gg t_m \tag{3}$$

During the final stage of annealing at 1173 and 1223 K, another anomaly of a sudden increase in the mean grain size is noticed (Fig. 5). This type of behaviour is indicative of a change in the growth mechanism from normal to abnormal. Abnormal grain growth has been reported [18, 19] to occur when some extraneous factors, such as second-phase particles or plastic strains, inhibit the normal process of growth. It is known that in this type of growth, only a few favourable grains grow at the expense of smaller ones, and the resultant microstructure depicts non-uniform grains [8]. The presence of a few large grains (Fig. 7) after annealing at 1223 K for 300 min is typical of such nonuniform grains observed in the present case. As exemplified by the mode of boundary migration in Fig. 8, grain-boundary pinning by second-phase particles such as TiC and Ti(CN) present in the steel [11, 12, 20], appears to be the primary cause of the abnormal growth. However, similar behaviour was not noticed when the steel was subjected to 1273 K and above, as the pinning effects of the carbides become sufficiently reduced due to high boundary mobility and increased diffusion rate of C and N. Based on Hillert's assertion [8], that normal grain growth is always preceded by abnormal growth, it is appropriate to assume that in the high-temperature range, abnormal growth stage is crossed much before the minimum 15 min annealing time studied.

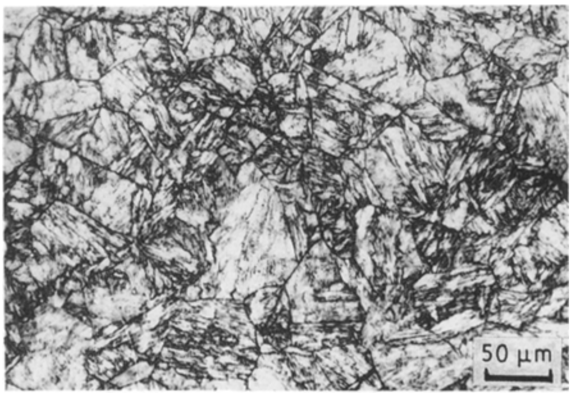

Figure 7 Non-uniform grains on annealing at 1223 K/300 min.

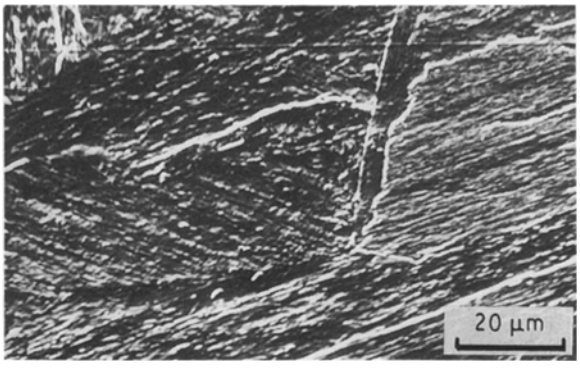

Figure 8 Scanning electron micrograph showing grain-boundary pinning.

3.2. Growth exponent

The growth exponent, n, calculated from the slope of the straight line portion of the log ΔD versus log t plot for both normal and abnormal grain growth, is given in Table II.

It is evident that up to 1273 K, the value of n remains fairly constant at 0.44 during the normal growth and is 2.0 and 0.9 for the abnormal grain growth at 1173 and 1223 K, respectively. While the observed n for the normal growth is acceptable for the steel processed through double vacuum melting, that obtained for the abnormal grain growth is in line with the value reported for pure Fe [21]. However, one significant deviation found in the present investigation is that the value of n decreases to 0.40 during normal growth at 1323 K. This is contradictory to the reported increase in n with increasing annealing temperature [7]. In order to understand this unusual behaviour, it is convenient to interpret the growth exponent from Equation (1) as

$$n = \frac{\mathrm{d}\Delta D/\mathrm{d}t}{\Delta D/t} \tag{4}$$

where $\mathrm{d}\Delta D/\mathrm{d}t$ is the instantaneous rate of increase and $\Delta D/t$ is the average rate of growth of the grains. The $\Delta D/t$ is expected to be faster at temperatures above 1323 K because the pinning effect is reduced by the coalescence of dispersed particles and high grain-boundary mobility. On the other hand, $\mathrm{d}\Delta D/\mathrm{d}t$ is impeded due to solute segregation at the grain boundaries. The evidence of Mo and Ti segregation in maraging steel has been extensively reported [22] and the effects of such segregation on the grain growth has been elaborately discussed by Leslie *et al.* [23]. It is because of the combined effects of these two factors that n in Equation 4 is reduced at 1323 K.

TABLE II Grain-growth exponent at different temperatures

Temperature (K)	Growth exponent, n	
	Normal	Abnormal
1173	0.44	2.00
1223	0.44	0.90
1273	0.43	—
1323	0.40	—

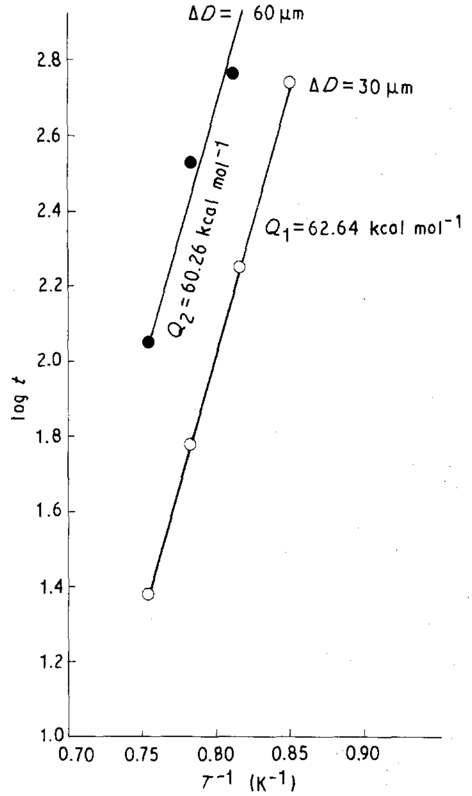

Figure 9 Activation energy for grain growth.

3.3. Activation energy

It is obvious from the preceding section that n remains fairly independent of temperature (except for the small decrease at 1323 K) during the normal growth and it is possible to show a time–temperature dependence of growth through the heat of activation. Fig. 9 is a plot of annealing time for a fixed increase in the grain size, ΔD, and the reciprocal of annealing temperature. The activation energies corresponding to 30 and 60 µm increase in grain diameter were found to be 60.0 and 62.5 kcal mol^{-1}, respectively. Because these values are in close proximity to the activation energy for the self diffusion of iron [24], it is inferred that the overall grain growth is controlled by self diffusion in iron. Similar observations were also made by Pepe and Savage [25] while studying the grain-growth behaviour in the heat-affected zone of the maraging steel welded samples. The activation energy in this case was reported to be 68 kcal mol^{-1}.

4. Conclusions

1. The grain growth in maraging steel follows the classical growth law of $D - D_0 = \Delta D = kt^n$.

2. A "time lag" was observed during the initial stage of annealing for the normal growth to set in, because of $\alpha - \gamma$ transformation resulting in a high density of dislocations.

3. Abnormal grain growth is observed after annealing at 1173 K for 480 min and at 1223 K for 300 min.

4. The growth exponent for the normal grain growth up to 1273 K is around 0.44, and for abnormal growth the values of n are 2.0 and 0.9 at 1173 and 1223 K, respectively.

5. The decreased value of n for the normal growth on increasing the annealing temperature to 1323 K is unique in this steel and results due to the combined effect of reduced instantaneous growth and increased average grain growth at this temperature.

6. The calculated activation energy in the range 60.0–62.5 kcal mol^{-1} indicates that the overall grain growth in this steel is controlled by self diffusion in γ-iron.

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