ISSN 0543-5846
METABK 44 (3) 169-174 (2005)
UDC - UDK 669.141.243:669–174:519.872.7:621.77=111

SECONDARY RECRYSTALLIZATION IN NON-ORIENTED ELECTRICAL STEELS

Received - Primljeno: 2004-06-23
Accepted - Prihvaćeno: 2004-10-25
Original Scientific Paper - Izvorni znanstveni rad

Modelling of kinetics of grain growth process after primary recrystallisation in different types of non-oriented electrical steels is discussed. The ferrite grain growth behaviour during secondary recrystallization was analysed by applying the general equation for grain growth. The activation energy for grain boundary motion in both semi-processed and fully processed steels was calculated. An idea of anisotropic mobilities is applied to the columnar grain growth description. It is shown that the value of activation energy for columnar grain development along progress normal direction is higher than the one for rolling direction.

Key words: *recrystallisation, activation energy, microstructure, rolling, electrical steel*

Sekundarna rekristalizacija u ne orijentiranim elektročelicima. Raspravljeno je modeliranje kinetike procesa rasta zrna nakon primarne rekristalizacije za različite vrste neorijetiranih elektročelika. Rast feritnog zrna tijekom sekundarne rekristalizacije analiziran je uporabom opće jednadžbe rasta zrna. Izračunata je energija aktivacije za gibanje granice zrna za poluobrađene i potpuno obrađene čelike. Za opis rasta stubičastih zrna korištena je ideja anizotropne pokretljivosti. Pokazana je da je vrijednost energije aktivacije za nastajanje stubičastog zrna veća u smjeru okomitom na smjer valjanja nego u smjeru valjanja.

Ključne riječi: *elektročelik, rekristalizacija, energija aktivacije, mikrostruktura, valjanje*

INTRODUCTION

Prediction of materials properties is connected with grain growth process. Grain size of the final microstructure is an important engineering parameter which influences the material properties such as fatigue, creep, yield strength etc [1-3]. It is important to control the final microstructure of electrical steels in terms of grain size and texture. The grain growth kinetics significantly affects the texture evolution and consequently the final magnetic properties of non-oriented electrical steels. Thus, modelling of grain growth is an important step in the production of this type of steels.

The aim of the statistical approach is to describe the grain growth process in order to predict the evolution of the grain size distribution, which classically represents the most comprehensive and experimentally accessible microstructural set of data. Such equations represent the basis for the statistical procedure, which is aimed on describing the grain size distribution trend. The common observation in metals and alloys is that the size distributions of grain aggregates during growth become equivalent when the measured grain size parameter R is normalised by the time-dependent average grain size $\bar{R}$. This means that grain structures are completely characterised, in a static sense, by simple probability functions of the standard deviation of the distribution together with the time dependence of average size scale $\bar{R}$ [4 - 6].

The general form of equation for grain growth is given as [7]:

$$\bar{R}^n - \bar{R}_0^n = ct \tag{1}$$

where: c and n are constants.

Secondary recrystallisation is an abnormal grain growth process where the driving force for boundary motion is generated by the boundary energy and “curvature effect” of growing grain. Abnormal grain growth occurs when the normal grain growth is stopped, i.e. a few grains rapidly coarsen thereby consuming smaller grains. The mean growth rate of grains of size R was postulated by Hillert [7]:

$$\frac{\mathrm{d}R}{\mathrm{d}t} = \alpha M \gamma \left(\frac{1}{R_\mathrm{c}} - \frac{1}{R} \right) \tag{2}$$

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where:

α - constant,
R_c - the critical radius at which grains neither shrink nor grow,
M - the mobility of a grain boundary,
γ - the grain boundary free energy per unit area.

In this contribution a theory and kinetics of grain growth in different types of non-oriented electrical steels will be discussed.

EXPERIMENTAL PROCEDURE

Both semi-processed (SP) and fully processed (FP1 and FP2) non-oriented electrical steels were used as experimental material (see Table 1.). SP samples were taken from the industrial process after cold and temper rolling processes and samples of FP - after cold rolling.

Generally, in order to obtain desired magnetic properties, non-oriented electrical steels are subjected to various annealing treatments in N_2-H_2-H_2O gas mixtures. In the case of semi-processed electrical steels the annealing is needed to relieve the residual stress and develop the grain growth and proper texture. In the case of fully processed electrical steels the annealing is performed to reduce the carbon content and to develop the grain growth with favourable texture. In the present investigation, the heat treatment was conducted in dry and wet atmospheres of cracked ammonia to investigate the secondary recrystallization process. The temperature of decarburization was varied in the range of 850 to 960 °C and the annealing time was changed from 1 to 45 min. During annealing, temperature was controlled within ±2 °C.

Table1. **Chemical composition of the investigated steels, in wt. %**

Tablica 1. **Kemijski sastav istraživanih čelika, mas. %**

Sample	FP1	FP2	SP
d / mm	0,65	0,67	0,65
C	0,031	0,004	0,05
Mn	0,38	0,375	0,36
Si	1,01	0,964	0,24
P	0,134	0,084	0,068
S	0,008	0,005	0,008
Al	0,157	0,159	0,11
N	0,007	0,005	0,005

The microstructure of the specimens was examined in the plane parallel to the sheet surface and in longitudinal cross-section using optical microscopy. The samples were polished and subsequently etched in 3 % Nital solution for 20 to 50 seconds in order to get all the grain boundaries visible. An average grain size was evaluated for each sample according to the procedure described in [8]. DIPS-5 image analysis software was used to make quantitative metallographic measurements. Grain size distribution was investigated by measuring the equivalent radius of approximately 1000 grains.

RESULTS AND DISCUSSION

Secondary recrystallization in non-oriented electrical steels is a special form of abnormal grain growth. This form of grain growth differs from typical abnormal growth in following important aspects:

1) the initial stages of abnormal growth are not sluggish (there is no a significant incubation period before grain coarsening);
2) abnormal grain growth in non-oriented electrical steels leads to wide grain size distribution (bimodal character of distribution was not proven);
3) secondary recrystallization occurs without a seletive growth process (in "classical" textured materials such as Fe-3%Si steels, the interaction between particles and grain boundaries is made in selective way; under the present investigation, grains of different orientation start to coarsen);
4) texture of final microstructure consists of several orientation components.

These facts suggest that grain growth in this type of steel is rather "intensive normal" than abnormal.

Many efforts have been made to simulate grain growth, either to confirm well-known theoretical assumptions or to compare simulation results with experimental data. Most of the grain growth theories [7, 9 - 11] lead to the parabolic grain growth kinetics:

$$d \sim t^m \tag{3}$$

where: m = 0,5.

The Monte Carlo simulation in [11] gives the grain growth exponent m close to 0,42. The fulfilment of this law was taken as one criterion in order to determine whether a simulation procedure is able to describe normal grain growth or not. Under the given investigation, the kinetics of grain growth is well described by the low of growth $d^2 \sim t^m$, with $n \approx 0{,}5$. The estimated values of exponent n presented in Table 2. are in disagreement with the ones presented for both normal and abnormal grain growth simulations [4 - 6, 9 - 11]. In [12] authors confirm that coefficient m in Equation (3) strongly depends on the annealing temperature. Under the present investigations these facts was not proven. The mentioned coefficient behaves as constant value in the investigated temperature range. A slight variation can be explained as an experimental error.

The "classical" abnormal grain growth rate increases with temperature according to the exponential relationship:

$$d^k \sim \exp\left(-\frac{1}{T}\right) \tag{4}$$

where: $k = 1$.

As will be shown below, the grain growth rate during secondary recrystallization in non-oriented electrical steels is described by exponential temperature dependence with $k = 2$ in Equation (2).

In order to make the interpretation of the results, compatible with an adequate accuracy in describing the main characteristic of abnormal grain growth process in non-oriented electrical steels, the ferrite grain growth behaviour during secondary recrystallization was analysed by applying the equation:

$$d^2 - d_0^2 = Ct^n \exp\left(-\frac{Q}{RT}\right) \qquad (5)$$

where:

d - the grain size at the given annealing time,
d_0 - the initial grain size,
t - denotes annealing time,
Q - the activation energy of grain growth,
T - annealing temperature.

The values of the coefficients C and n depend on the type of material. Using Equation (5), the value of activation energy Q can be found from the dependence of $(d^2 - d_0^2)$ on the $(1/T)$ in semi-logarithmic scale for the fixed annealing time.

Table 2. **Coefficients of the grain growth kinetic in the investigated non-oriented electrical steels**
Tablica 2. **Koeficijenti kinetike rasta zrna u istraživanim neorijentiranim elektročelicima**

Sample	n	Temperature range / °C
FP1	0,48	890 - 950
FP2	0,49	780 - 960
SP after TR	0,45	780 - 880
SP without TR	0,46	780 - 880

In spite of the fact that a complete and exact modelling of grain growth phenomena is complicated by a number of different factors, the simulation with some simple assumption may lead to profound insight into the mechanism of processes. The development of this semi-empirical approach is based on the experimental observations and above mentioned principles of grain growth in non-oriented electrical steels.

Table 3. **The values of activation energies connected with ferrite grain growth [4]**
Tablica 3. **Vrijednosti energija aktivacije koje se odnose na rast feritnog zrna [4]**

Process	Q / (kJ/mol)
Volume diffusion of Fe in α iron	280
Grain boundary diffusion of Fe in	164
Grain boundary mobility of pure iron α iron	147

The activation energy of secondary recrystallization for grain boundary motion was calculated in semi-processed steel SP (Figure 1.a). Because of low concentration of alloying elements, the value of Q = 145 kJ/mol (Figure 1.a, line 2) is comparable with the activation energy for grain boundary mobility in pure iron (see Table 3.). The activation energy Q = 91 kJ/mol for the grain growth of the temper rolled steel (Figure 1.a, line 1) is lower than the activation energies for processes associated with ferrite grain growth. Comparing value of Q in temper rolled material to the corresponding value of the same material without temper rolling (see Figure 1.a) make it possible to claim that temper rolling means additional driving force for ferrite grain

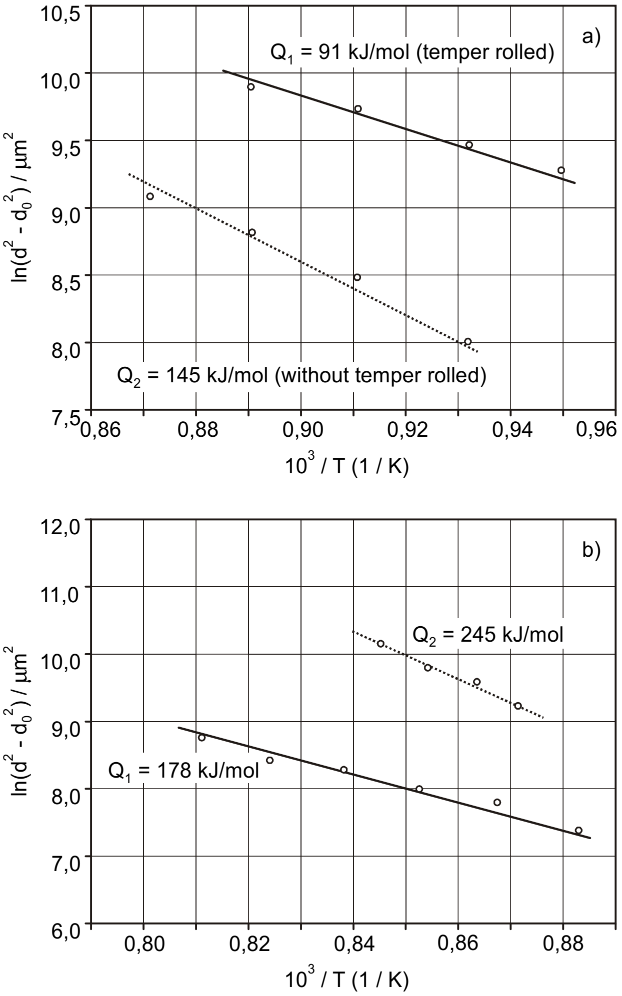

Figure 1. **Plot of $\ln(d^2 - d_0^2)$ as function of $1/T$ for ferrite grain growth in: a) steel SP; b) steels FP2 (line 1) and FP1 (line 2)**
Slika 1. **Prikaz ovisnosti $\ln(d^2 - d_0^2)$ o $1/T$ za rast feritnog zrna: a) čelik SP; b) čelici FP2 (linija 1) i FP1 (linija 2)**

growth. The observation that a small prior plastic strain may promote anomalous grain growth is widely known [7] and interpreted as strain induced grain boundary migration. Deformation leads to two contributions in microstructure changes i.e. increase of dislocation density and grain elongation with an associated increase of grain boundary area. Furthermore, the stored energy of cold rolled material depends on the local orientation of grains. The difference in stored energy provides the driving force for strain induced grain boundary motion that can lead to the preferential formation of recristallized nuclei in low stored energy regions. These facts support the suggestion that a non-equilibrium grain boundary motion is induced by plastic deformation, i.e. by temper rolling, and as a result, material shows higher mobility of grains in comparison with non-deformed material.

The polygonal microstructure creation (Figure 2.a) in ultra low carbon steel FP2 is connected with the relatively high value of activation energy $Q \sim 178$ kJ/mol (Figure 1.b). The latter is comparable with the activation energy for grain boundary diffusion of Fe in ferrite (see Table 3.). The activation energy for grain growth in fully processed steel FP1 (Figure 1.b) was estimated as ~ 245 kJ/mol (Figure 1.b). The calculated value is comparable with self-diffusion of Fe in ferrite (see Table 3.).

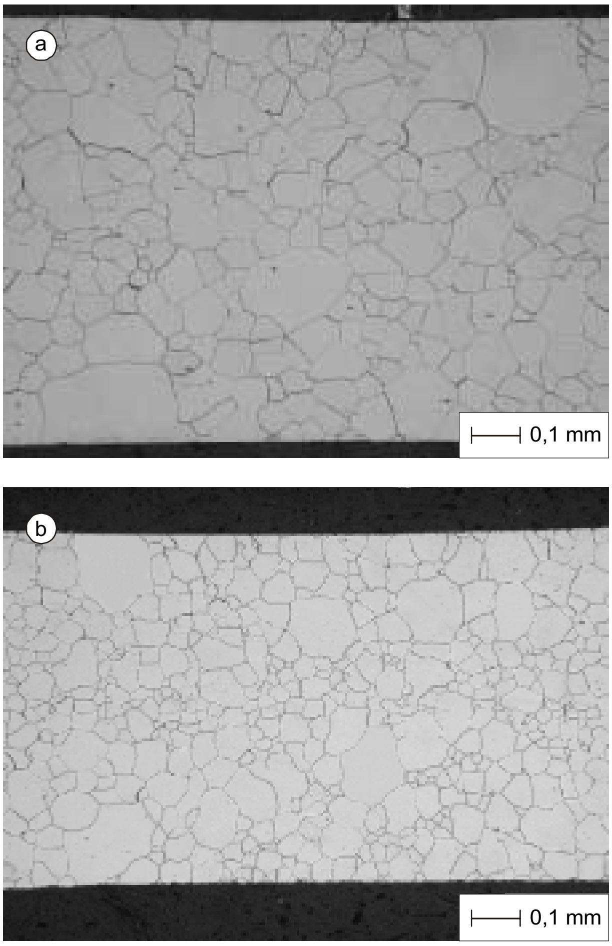

Figure 2. **Microstructures of the investigated samples annealed at 875 °C for 5 min in wet NH_3 atmosphere: a) steel FP2; b) steel FP1**

Slika 2. **Mikrostrukture uzoraka istraživanih čelika odžarenih kod 875 °C u vremenu od 5 min u vlažnoj NH_3 atmosferi: a) čelik FP2; b) čelik FP1**

The chemical composition of both FP1 and FP2 steels is similar, apart from carbon content. Carbon addition to laminate steels decreases the α to $\alpha + \gamma$ transformation temperature. The temperature Ac_1 was calculated by software "thermo-calc" for FP1 and FP2 as 880 °C and 962 °C respectively. It is obvious that phase transformation does not occur in steel FP2 in the investigated temperature range. By comparing activation energy of steel FP1 for grain growth with the corresponding activation energy of steel FP2 (see Figure 1.b) one can conclude that the ferrite grain growth without phase transformation needs lower activation energy than the one with phase transformation. Furthermore, carbon in solid solution, which is preferentially enriched at the moving grain boundary, reduces grain boundary mobility of the growing grain due to a solute drag effect. Thus, difference in chemical composition leads to distinct mechanisms of grain boundary motion, as a consequence of different phase composition at elevated temperature.

Recently, a significant amount of experimental work has been carried out on steels to tailor the properties. A common difficulty in many cases is the inhomogeneity of the microstructure and the texture of steel as a result of the production process. This is of particular importance in electrical steels. For low carbon steel it is possible to apply a decarburizing annealing in the intercritical region that leads to the columnar-grained microstructure with a specific type of the texture [14]. The columnar grain growth of fully processed steel FP1 is driven by diffusion-induced grain boundary motion mechanism. The moisture of gas mixture, temperature and heating rate is the main parameters that influence the directional grain growth [14]. Carbon diffusion along the grain boundary presents an elastic stress on boundary that causes motion of latter. A discontinuous concentration profile of carbon is observed during decarburization [13]. The later is caused by the continuity of chemical composition of carbon in ferrite and austenite. The carbon content increases from zero at the surface to a quasi-equilibrium value at the $\alpha / (\alpha + \gamma)$ interface. Thus, decarburization proce-eds through "diffusion pipes" that are aligned with normal direction (ND) to the sheet-plane. The blocking of carbon in the "diffusion pipe" decreases the rate of carbon removing and, thus, decreases the elastic stress along the grain boundary. Then, the retarded decarburization produces high pinning pressure on grain boundary. For optimal columnar grain growth during the decarburization, the carbon removing should have a sequential character. It is well known that the boundary velocity is proportional to the frequency of atoms "jump" across the grain boundary plane. On other hand, the intensive decarburization should accelerate "jumping process" of matrix atoms along the normal direction. Then, intensive decarburization causes small pinning effect on grain boundary that leads to directional grain boundary motion.

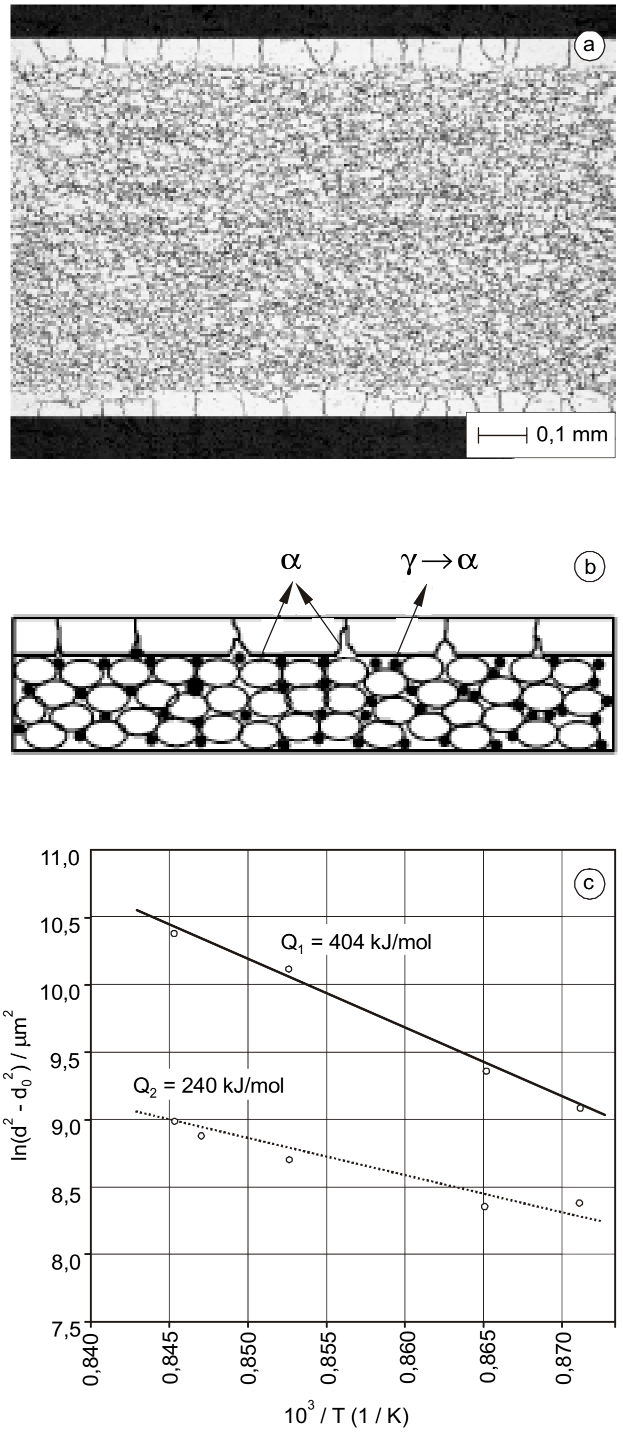

Figure 3. **a) Columnar-nucleate microstructure at the commence of decarburization process in steel FP1; b) Scheme of columnar microstructure creation; c) Plot of ln(d^2- d_0^2) as function of 1/T for columnar-nucleate creation along normal direction (line 1) and rolling direction (line 2) in steel FP1**

Slika 3. **a) Nukleacija stubičaste mikrostrukture na početku procesa razugljičenja za čelik FP1; b) Shematski prikaz nastajanja stubičaste mikrostrukture; c) Prikaz ovisnosti ln(d^2 - d_0^2) o 1/T za nukleaciju stubičaste mikrostrukture za čelik FP1 u smjeru okomitom na smjer valjanja (linija 1) i u smjeru valjanja (linija 2)**

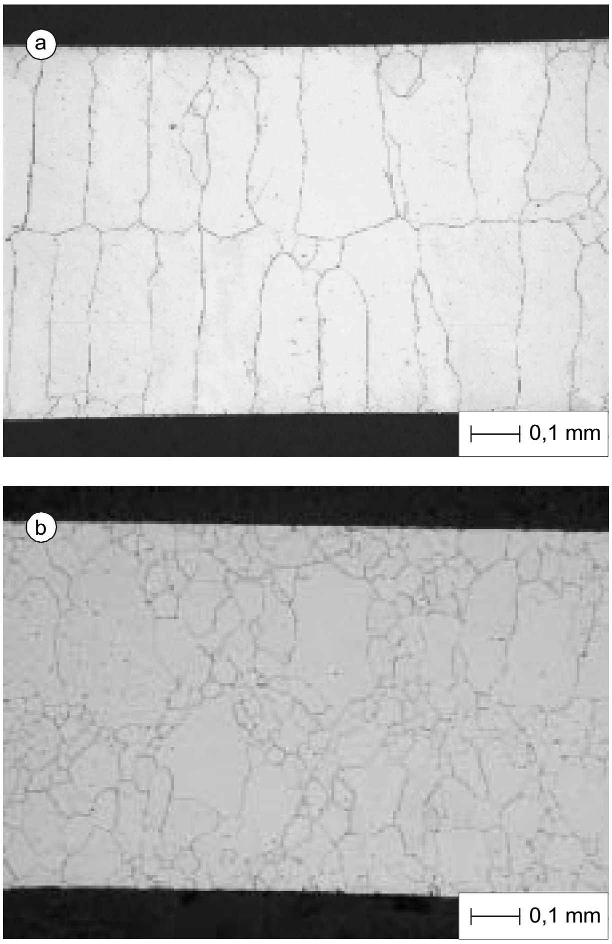

Figure 4. **a) Columnar microstructure of steel FP1after annealing when the activation energy of columnar-nucleate creation is $Q \sim 400$ kJ/mol; b) Microstructure of steel FP1after annealing when the activation energy at the beginning of process is $Q \sim 280$ kJ/mol**

Slika 4. **a) Stubičasta mikrostuktura čelika FP1 nakon žarenja kod energije aktivacije Q ~ 400 kJ/mol; b) Mikrostruktura čelika FP1 nakon žarenja kod energije aktivacije na početku procesa Q ~ 280 kJ/mol**

As the microstructure is non-equiaxed the activation energy of grain growth may be considered as dependent value on growth direction. In the field of grain growth simulation, the Monte Carlo algorithm is widely used to model anisotropic grain growth [7]. In the present work, the idea of anisotropic mobilities is applied to the columnar grain growth description. The activation energy was calculated for two growth directions, i.e. rolling direction (RD) and normal direction (through thickness). When material is heated at proper rate, at the temperatures lower than Ac_1

primary recrystallization occurs simultaneously with intensive decarburization of the surface material layers. After material is heated up to temperatures above Ac_1, phase transformation, which refines primary recrystallized matrix, occurs in the inside region. At the same time, surface layers are not subjected to the phase transformation because of the low local concentration of carbon. First, the small g phase grains are transformed into a ones and as a result the material has a fine primary recrystallized microstructure and the thin decarburized a-region on the sheet surface (Figure 3.a, b). Figure 3.c describes the formation process of columnar microstructure from the point of activation energy. From this figure, the values of Q for grain growth were estimated as 240 kJ/mol and 404 kJ/mol for RD and ND respectively. The activation energy for columnar grain growth in the plane of RD is comparable with activation energy of self-diffusion of Fe in ferrite. The high value of Q for progressive ND (404 kJ/mol) confirms that columnar grain growth needs high energy to start the direct abnormal growth, and this can be explained by the energy demand for phase transformation during heating and subsequent growth of formed α grains on the surface. At later stages of this process, the size advantage of surface ferritic grains in comparison with fine-grained inner material is high and enough for continuation of directional abnormal grain growth of surface layer towards midplane. Coarsening of grains in the midplane is very slow because of the high “activation energy barrier”. After finishing the decarburization process, the microstructure is fully ferritic and grains have columnar shape (Figure 4.a). As the value of Q (at the beginning of the process) is lower than the critical activation energy for columnar nucleate creation, the inhomogeneous microstructure development takes place (Figure 4.b).

SUMMARY

Depending upon the chemical composition of the steels under the investigation, the polygonal grain growth is linked with either the grain boundary diffusion or the self-diffusion of Fe in ferrite.

By applying the temper rolling process, the activation energy for grain growth can be lowered during the secondary recrystallization. This fact may suggest that a non-equilibrium grain boundary motion is induced by plastic deformation and as a result, material shows higher mobility of grains in comparison with non-deformed material.

A model of anisotropic mobilities is applied to the columnar grain growth description. The value of activation energy of grain growth, for early stage of columnar microstructure development, along progress normal direction is higher than the one for rolling direction. The high value of activation energy for grains propagation with normal to sheet plane direction towards sheet midplane (Q ~ 404 kJ/mol) confirms that columnar grain needs high energy to start and follow the directional abnormal growth.

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