Microstructural Characterization and Recrystallization Kinetics Modeling of Annealing Cold-Rolled Vanadium Microalloyed HSLA Steels

Zhenyu Liu, Raymundo Ordonez Olivares, Yinkai Lei, Calixto Isaac Garcia, and Guofeng Wang*

Department of Mechanical Engineering and Materials Science, University of Pittsburgh,
Pittsburgh, PA 15261, USA

* Corresponding Author: guw8@pitt.edu

Abstract

In this study, we investigated the microstructural evolution and recrystallization kinetics during the annealing of cold-rolled Vanadium microalloyed HSLA (denoted as HSLA-V) steels using both experimental characterization techniques and theoretical model analysis method. Our experimental results indicate that the recrystallization kinetics can be quantified with the evolution of volume fraction of recrystallization as a function of annealing time, and is strongly dependent on the processing parameters (i.e., cold-rolling reduction and annealing temperature) of the HSLA-V steels. Validated by experimental data, our physics-based model analysis revealed that the recrystallization process in the HSLA-V steels consisted of an early stage with both nucleation and grain growth processes and a later stage with grain growth only. Moreover, the growth of the recrystallized grains was found to follow one dimensional preferential growth mechanism, owing to the dispersed V(C,N) precipitates, in the HSLA-V steels. Consequently, our new analysis model is demonstrated to be useful to gain insights into the recrystallization kinetics in the annealed cold-rolled HSLA-V and even other microalloyed steels.

Keywords: metals and alloys, kinetics, microstructure, Electron Back-Scattering
Diffraction, thermodynamic modeling

1. Introduction

High-strength low-alloy (HSLA) steel is a special category of plain carbon steels with microalloying elements, such as vanadium (V), niobium (Nb), or titanium (Ti), and possesses superior mechanical properties [1, 2]. The yield strength of some HSLA steels can reach as high as 690 MPa, which is more than twice higher than the strength (in the range 170-250 MPa) of typical plain carbon steels [1-4]. The high strength of HSLA steels is believed to stem from their microstructural factors such as grain refinement [5, 6], precipitation hardening [7, 8], and inclusion shape control [9, 10]. As a specific example, V-microalloyed HSLA (denoted as HSLA-V) steels with ultrafine grains have been found to exhibit an excellent balance of strength and ductility [11, 12], and consequently, have been employed in a wide range of applications (for examples, bridges, buildings, vehicles, and pipes) where materials reliability, environmental issues, and fabrication costs are main factors of design considerations [13].

In many applications, HSLA steels are cold-rolled into sheets first in order to fabricate a variety of final product components. However, cold rolling process could exert detrimental influence on the microstructure and mechanical properties of the cold-rolled steels. Large amount of dislocations are generated inside the steels during cold rolling process. These dislocations would pile up along their preferred slip planes and cause undesired work-hardening in cold-rolled steel sheets. Moreover, cold rolling could lead to extension of grains along the rolling direction, i.e., directional microstructural texture, and thus anisotropic mechanical properties in the processed steels [14, 15]. Therefore, a subsequent annealing process is normally needed to remove residual stress, reduce anisotropy of mechanical properties, and acquire equiaxed fine grains of the cold-rolled HSLA steels. A previous study demonstrated that an annealing process at 600°C for 1 hour could fully recover the optimized mechanical properties of the cold-rolled HSLA steel which had undergone above 60% deformation [16].

Therefore, it is of great technological and scientific interests to attain a physics-based

kinetics model that can reliably and efficiently predict the effect of subsequent annealing on the microstructural characters of cold-rolled HSLA steels. To this end, we present here both the experimental characterization of microstructure evolution and the theoretical analysis of recrystallization kinetics during the annealing of HSLA-V steel with severer cold-rolling deformation. In particular, we focus this study on elucidating the relation between processing parameters (including extent of cold-rolling deformation, annealing temperature, and annealing time) and resultant microstructure (characterized by recrystallization volume fraction) of the HSLA-V steels investigated.

2. Materials and Methods

2.1. Materials and experimental procedures

The as-received steel in this study is a commercial V-bearing high strength low alloy HSLA-V steel obtained from NUCOR Berkley. The steel had been previously hot rolled and coiled at 650°C to produce a hot band condition. The average chemical composition of the as-received steel is given in Table 1. We further processed the steel with cold rolling using a laboratory rolling mill at room temperature. During the cold rolling, the lateral expansion of the material was restricted in order to preserve plain strain condition. It is worth mentioning that we found that the microstructure inhomogeneity through the wall thickness of the cold-rolled sheets was insignificant when the cold rolling deformation was above 35% reduction. To minimize the influence of deformation inhomogeneity on our results, we conducted the study in this work for three different types of full-hard steels attained after deforming the hot band steel supplied, as sheets of approximately 6.18 mm thickness, by 40 %, 60 % and 80 % reduction under the plane strain condition. Following the cold rolling, the steels were annealed at three different temperatures (630 °C, 650 °C, and 670 °C) and for various annealing duration (from 15 min to 720 min), with an averaged heating rate of 8.6 °C/min and cooling rate of 52.3 °C/min. Samples from the materials in the as-received and as-processed condition were sectioned, mounted and prepared

according to well-accepted metallographic techniques (as shown schematically in Fig. 1) [17]. In this study, the samples were mounted onto special Cu-compound substrates and polished using vibration polishing/electro-polishing machines. The Vibromet 2 Vibratory Polisher (BUEHLER) is used for fine polishing. The total sample polishing time varies from 4 to 6 hours depending on sample hardness until desired surface condition is obtained.

The microstructure of the prepared HSLA-V steel samples were characterized by an optical Keyence VHX 5000 microscope and a Philips XL 30 field emission gun scanning electron microscope (FEG-SEM) equipped with an electron back-scattering diffraction (EBSD) analyzer. In this work, we selected the mid-intersection surface perpendicular to the normal direction for the EBSD characterization.

Figure 1. Schematic diagram depicting sample preparation and mounting procedure employed in this study. In the figure, symbol ‘RD’ stands for ‘Rolling Direction’, symbol ‘ND’ stands for ‘Normal Direction’, and symbol ‘ δ ’ refers to the thickness of cold rolled steel samples.

Table 1. Chemical composition of the HSLA-V steel used in this study.

	C	S	P	Mn	Si	Ti	Cr	Mo	V	Al	Fe
wt.%	0.06	<0.002	<0.008	0.80	0.025	0.002	0.088	0.010	0.06	0.025	Bal.

2.2. Numerical analysis approach

In this work, we described the transformation kinetics of the recrystallization process in the annealing cold-rolled HSLA-V steels using an improved Johnson–Mehl–Avrami–Kolmogorov (JMAK) model which assumes that the nucleation of the recrystallized grains is homogeneous and the growth of the recrystallized grains is isotropic in the deformed steels [18-21]. According to the JMAK model, the degree of transformation f (defined as the ratio of transformed (i.e., recrystallized) volume to the total volume) at annealing time t is expressed as

$$f = 1 - \exp\left(-g \int_0^t I_v \left(\int_{t'}^t u d\tau\right)^m dt'\right) \quad (1)$$

In this equation, I_v is the nucleation rate per unit volume, u is the grain growth rate, g is the geometry factor, m is the growth dimension, and t' is the time at which a nucleus of recrystallized grain forms. By numerically fitting the experimental recrystallized fraction measured in our experiments into the JMAK model, we were able to extract transformation kinetic information (such as the grain growth dimension and total activation energy) for the recrystallization process in the cold-rolled and subsequent annealed HSLA-V steels.

3. Results and Discussion

3.1. Microstructure characterization and analysis

Figure 2 shows the microstructural feature of our samples in three different processing conditions etched with 2% Nital. Fig. 2(a) displays a typical hot band microstructure, in which ferrite (bright phase) and cementite carbides and pearlite (the black agglomeration inside the grains and along ferrite grain boundaries) are distinguishable, in the as-received

steel. Due to the nature of the etching process the black lines in optical microscopy correspond to chemical differences (ferrite and pearlite), crystallographic texture of the grains, and/or energy differences in the grain boundaries. In Fig. 2(b) one can observe the elongated grains along the rolling direction (horizontal direction of the photo) in the full-hard sample obtained after cold rolling by 60% reduction; whereas Fig. 2(c) shows a more equiaxed microstructure, with more rounded grains, in the recrystallized sample after heat treated at 650°C for 12 hours. Consequently, the recrystallization in the annealed HSLA-V steels occurs in the form of new equiaxed grains replacing elongated grains to reduce stored deformation energy. Hence, we can use the fraction of the grains transformed from elongated to equiaxed shape as an empirical parameter to quantify the extent of recrystallization in our annealed samples.

Figure 2. Optical microscopy images showing the microstructure of the HSLA-V steels in the (a) as-received (hot-rolled), (b) full-hard (60% cold-rolled) (c) recrystallized conditions (annealed at 650°C for 12h).

In this study, we employed standard operation procedure of EBSD observation to determine the stored deformation energy and the volume fraction of recrystallization in our cold-rolled and annealed steel samples. In every EBSD observation of this study, we used 20 kV acceleration voltage, a beam spot size of 6, a square scan mode, and a scanning step size from 0.1 μm to 0.5 μm , depending on grain size. All the collection and analysis of EBSD data were conducted using the TSL OIM data collector software and the TSL OIM Analysis software, respectively. The software developed by Choi [22] was employed to analyze the stored energy distribution on each sample.

The stored energy in the HSLA-V samples is extracted from EBSD analysis on the observed subgrains in the deformed microstructure. This analysis method is a new way to quantify the heterogeneous distribution of stored energy in deformed metal materials and works as follows. Provided that the dislocation substructure in the deformed HSLA-V sample can be simplified to be in equivalent to subgrain (i.e., grains bound with low angle grain boundaries) with diameter D and boundary energy γ , the stored energy (SE) of each lattice site can be written as [23]:

$$\text{SE} = \alpha \frac{\gamma}{D} \quad (2)$$

where α is a geometric constant (we assume its value to be 3 in this study). Further, the energy of a low-angle boundary is dependent on the degree of misorientation between the neighboring grains up to the transition to high-angle status. In the case of simple tilt boundaries the energy of a boundary made up of dislocations with Burgers vector b and spacing h is predicted by the Read-Shockley equation [23]:

$$\gamma(\Delta\theta) = \begin{cases} \gamma_m \frac{\theta'}{\theta^*} \left[1 - \ln \left(\frac{\theta'}{\theta^*} \right) \right], & \theta' < \theta^* \\ \gamma_m, & \theta' \geq \theta^* \end{cases} \quad (3)$$

where $\theta' = |\Delta\theta|$ if $0 \leq |\Delta\theta| \leq \pi$ and $\theta' = 2\pi - |\Delta\theta|$ if $\pi \leq |\Delta\theta| \leq 2\pi$, $\Delta\theta$ is taken as the boundary misorientation between two neighboring subgrains, θ^* is the misorientation limit for low angle grain boundaries, which was chosen to be 15° in this

study [23], and γ_m is the specific energy of high angle grain boundaries. In this study, the value of γ_m was chosen to be 835 mJ/m², which is the suggested value for stainless steels in the literature [22]. Taking the grain boundaries with all possible misorientation angles into computation, we plot in Fig. 3 some representative stored energy maps of our cold-rolled and annealed samples. These maps were made by using the abovementioned Subgrain Method to evaluate the orientation-dependent stored energy of deformed grains from EBSD measurements. It can be observed in Fig. 3 that the stored energy in the sample decreases continuously with annealing time. Our quantitative measurements show that the stored energy in the cold-rolled sample would be reduced by 52.4% after the first 15-minute annealing and be nearly depleted (reduced by 86.8%) after annealed for 12 hours. More importantly, our results in Fig. 3 revealed clearly that the decrease in the stored energy during annealing was due to the nucleation and growth of recrystallized grains (with no deformation) in the sample.

Furthermore, we determined the volume fraction of recrystallization by analyzing the Kernel average misorientation (KAM) distribution in our polycrystalline samples [24]. In this study, the Kernel average misorientation distribution maps were attained by processing the original EBSD scanning data with the TSL OIM analysis software. In the analysis, the misorientations between the center point of a grain and all the points at the perimeter of the kernel were measured and then a local misorientation value (i.e., the average of all these misorientations) was assigned to the center point. It should be noted that we excluded all the misorientations larger than 5° in our average computation for elimination of any contribution from high angle boundaries. Figure 4 shows the Kernel average misorientation distribution maps determined from our EBSD data to quantify the fraction of recrystallization in an annealed HSLA-V sample. In Fig. 4, the yellow or red regions represent the grains having high KAM values and hence high dislocation density whereas the green or blues regions mark the grains having low KAM values and low dislocation density. Our results in Fig. 4 show clearly the process in which the deformed grains (yellow,

red and green regions) from cold-rolling gradually disappear and new deformation-free, recrystallized grains (blue regions, where the KAM parameters are of values less than 0.7°) emerge and grow with annealing time. It should be pointed out that our choice of the KAM threshold (0.7°) for recrystallized grains is the median of the typical values (ranging from 0.5° to 1.0°) used in the EBSD studies of recrystallization in steels [24-27]. Consequently, we can quantitatively determine the volume fraction of recrystallization in our samples at different annealing stages by measuring the fraction of blue regions in our KAM distribution maps. For each sample, we conducted one EBSD analysis on an area of $80 \times 80 \mu\text{m}^2$ with scanning step size of $0.1 \mu\text{m}$ to $0.5 \mu\text{m}$. Hence, the volume fraction of recrystallization in our sample was determined to be the average value over 25,760 to 640,800 scans.

Figure 3. Measured stored energy distribution of (a) a cold-rolled sample (60% reduction) and (b-f) annealed samples after various annealing times at 650°C from EBSD analysis. The dark contrasts in the figure are the substructures, consisting mainly of high angle grain boundaries, without recrystallization in the sample.

Figure 4. Kernel average misorientation distribution maps of the HSLA-V steel in (a) cold rolled (full-hard) condition, and after annealed at 650°C for (b) 60 minutes and (c) 720 minutes. In the figure, color blue, green, yellow, and orange and red represent the kernel average misorientation in the range of (0°, 0.7°), (0.7°, 1.4°), (1.4°, 2.1°), (2.1°, 2.8°), and (2.8°, 3.5°), respectively.

3.2. Model Process Kinetics

In a conventional JMAK model, the degree of transformation f (i.e., volume fraction of recrystallization) is predicted to have a function form of $f = 1 - \exp(-kt^n)$ in term of annealing time t , under the assumption that nuclei are formed at a constant rate and grains also grow at a constant rate. Hence, quantity $\ln(-\ln(1-f))$ should exhibit a linear relation with respect to $\ln(t)$. However, our experimental data for the HSLA-V steel samples cold-rolled by 60% reduction and subsequent annealed at 650 °C (inset of Fig. 5)

show that $\ln(-\ln(1-f))$ has a linear relation with $\ln(t)$ with an Avrami exponent of 1.72 below annealing time of 1 hour, whereas with an Avrami exponent of 0.57 above annealing time of 1 hour. Apparently, the recrystallization kinetics in the HSLA-V steels deviates appreciably from the conventional JMAK model. Hence, some modifications to the JMAK model are needed in order to properly describe the recrystallization process in the HSLA-V steels annealed.

This phenomenon of a decreasing growth rate of recrystallization was often attributed to recovery process [28, 29] and/or the inhomogeneity of the stored energy [30, 31] in the samples. In specific for steels, some studies suggested that the recovery process only played a minor role but the heterogeneous distribution of the stored energy was the main reason underlying the non-linear JMAK plots [32, 33]. Oyarzábal et al [34] have performed extensive characterization study on the effect of the stored energy and recovery on the recrystallization kinetics in a cold-rolled low carbon steel, which exhibited clearly a two-range recrystallization kinetics similar to one shown in Fig. 5. In the study, they found that both nucleation and grain growth took place in the early range whereas the number density of the recrystallized grains remained almost constant at the later range of the recrystallization process. Oyarzábal et al [34] also confirmed that recovery made only slight contribution to the decrease in the total stored energy in the samples. We believe that the recrystallization kinetics in our HSLA-V steels follows the similar one described by Oyarzábal et al in Ref. [34].

Figure 5. Schematic diagram showing the main features of our modified JMAK model for recrystallization kinetics. Before t_s , the recrystallization process involves both nucleation and grain growth; after t_s , the recrystallization process involves only grain growth. The inset figure shows the similar plot of experimental values obtained from the HSLA-V steel samples cold-rolled by 60% reduction and subsequent annealed at 650 °C.

Consequently, we assume that the whole recrystallization process consists of two stages in our modified JMAK model (schematically shown in Fig. 5). In an early stage of the recrystallization process (below annealing time t_s), both nucleus formation and grain growth could take place following the conventional JMAK model. However, only grain growth from the early recrystallized grains could occur (namely, nucleation completely stops) in a later stage of the recrystallization process (above annealing time t_s). This assumption is similar to that of the site saturation model [23]. We believe that the aforementioned assumption is in better consistence with our experimental observation for the HSLA-V steels. In Fig. 6, we show how the measured stored deformation energy in an annealed HSLA-V sample varies with respect to annealing time. It can be noticed in Fig. 6 that the stored energy decreases rapidly in the early stage of the recrystallization (roughly

speaking, below 1 hour) but decreases quite slowly in the later stage of the recrystallization. It is conceivable that the nucleation of recrystallized grains requires some critical stored energy as driving force to overcome the energy barrier associated with the generation of interphases between the recrystallized nucleus and the unrecrystallized materials. Therefore, we introduced a tunable parameter, t_s , below which the nucleation of recrystallized grains occurs at a constant non-zero rate but above which the nucleation rate is zero, in our modified JMAK model.

Figure 6. Evolution of stored deformation energy with annealing time in the HSLA-V steels that was cold-rolled by 60% reduction and then annealed at three different (630°C, 650°C, and 670°C) temperatures. In the figure, the circles represent our experimental values measured using EBSD method and the lines show function $SE = a * \exp(-b * t^c) + d$ with parameters a , b , c , and d best fitted into the experimental data at each annealing temperature.

Moreover, we modified the growth rate term in the JMAK model in this study. Instead of using the typical constant growth rate approximation [35], we assume that the growth of the recrystallized grains in the annealed HSLA-V steels follows a parabolic time law and

thus the averaged grain size is proportional to function $\sqrt{t}$ with an evolution of annealing time, by analogy with the parabolic ferrite growth law in austenite-ferrite transformation [36]. Namely, $\int_{t'}^t u d\tau = A\sqrt{t - t'}$, here A is a proportional coefficient and t' is the time when the recrystallized grain starts to grow. We can also justify our modified assumption here using our experimental data. It can be seen in the inset of Fig. 5 that the slope of the linear relation between $\ln(-\ln(1-f))$ and $\ln(t)$ is smaller than 1.0 for our HSLA-V steel samples. This result suggests that the grain growth rate must be slower than a constant rate growth during the recrystallization process of the HSLA-V steels. Our choice of the parabolic growth rate is stemmed from the postulate that the grain growth during recrystallization is related to high angle boundary migration [35, 37] and hence is rate-controlled by grain boundary diffusion process [38].

In summary, the recrystallization kinetics in the HSLA-V steels which were cold-rolled and then annealed can be described as follows in our modified JMAK model.

$$\begin{aligned} f &= 1 - \exp\left(-C \frac{2}{m+2} t^{\frac{m+2}{2}}\right), & \text{when } t < t_s \\ f &= 1 - \exp\left(-C \frac{2}{m+2} \left(t^{\frac{m+2}{2}} - (t - t_s)^{\frac{m+2}{2}}\right)\right), & \text{when } t > t_s \end{aligned} \quad (4)$$

In this equation, $C = gIvA^m$ is a parameter related to the rates of nucleation and grain growth, A is the proportional coefficient of grain growth rate, m is a parameter equivalent to Avrami exponent and related to grain growth dimensions, and t_s is a tunable parameter indicating when the nucleation process stops. It is noted that both C and t_s depend strongly on the degree of deformation (i.e., volume reduction in our case) from cold-rolling and the temperature of subsequent annealing, whereas m should be insensitive to these processing parameters.

3.3. Model analysis results

In this study, we used the model (Eq. (4)) best fitted into the experimental data (specifically, the measured recrystallized fraction as a function of time) to extract the kinetic information about the recrystallization process of the cold-rolled HSLA-V steels.

Figure 7. Best model fittings (assuming $m=1$, 2, or 3) to the experimentally measured recrystallization fraction for samples with 60% cold-rolled reduction and annealed at 650°C.

In Eq. (4), m is a parameter equivalent to Avrami exponent and related to grain growth dimensions. In Fig. 7, we show our best model fittings against a set of representative experimental results with the assumed value of m to be 1, 2, and 3, respectively. It was found in Fig. 7 that our model would significantly underestimate the recrystallization fractions at early recrystallization stage (short times) but overestimate the recrystallization fractions at late recrystallization stage (long times) when m assumed values of 2 or 3. In contrast, when m assumes value of 1, our model yields an excellent agreement (coefficient of determination, $R^2=0.9886$) with the experimental results of the variation of the recrystallization fraction as a function of annealing time in the cold-rolled HSLA-V steels.

These results suggest that the recrystallization process in the cold-rolled HSLA-V steels proceeds with a growth dimension m of 1 and hence an Avrami exponent of 0.5 at the grain growth stage. Our results are in consistent with previous results reported by Garcia-Mateo et al. [39] that the Avrami exponent should be around 0.5 in warmed worked vanadium microalloyed steels recrystallized at 835 °C. It has been proposed that the recrystallization

mechanism underlying an Avrami exponent of 0.5 is a preferential growth of the grains with some particular orientation owing to precipitation phases in the steels. In an analogous example, it was found that the $\{111\}$ -oriented grains would grow faster than the grains with other orientations due to the drag effect of AlN particles in the recrystallized Aluminum-killed steels [40]. In the V microalloyed steels, it has been known that the decomposition of austenite to ferrite is accompanied by the formation of carbide precipitation along the interfaces [7, 41]. Specifically, Shanmugam et al [42, 43] observed the cuboid (45-70 nm), spherical/irregular (20-45 nm), and fine/needleshaped V(C,N) precipitates formed in the V microalloyed steel hot-rolled and coiled at around 600°C. Hence, we believe that the drag effect of these interphase carbide precipitations (such as V(C,N) particles) is responsible for the observed one dimensional growth of the recrystallized grains in the HSLA-V steels. Consequently, we set the value of m to be 1 in our further analysis of the process kinetics in the annealing cold-rolled HSLA-V steels.

In Fig. 8, we plotted the best fit of our recrystallization kinetics model (Eq. (4)) to the recrystallization rate measured in the HSLA-V steels which underwent various cold-rolling reduction (i.e., 40%, 60%, and 80 %) and subsequent annealing at different temperatures (i.e., 630 °C, 650 °C, and 670 °C). In all the cases, our model curves agree well with the experimental data through the whole recrystallization process. Especially, for the HSLA-V steels with 60% and 80% cold-rolling reduction, our model gives a fine description of the experimental data with coefficient of determination R^2 all greater than 0.975.

Moreover, we presented the recrystallization kinetic parameters of the HSLA-V steels attained from our data fitting in Table 2. Our results for the HSLA-V steels in Table 2 show that recrystallization kinetic rate C (which includes both the nucleation and growth rates) increases appreciably with an increase in the cold-rolling reduction and annealing temperature, indicating that both the stored strain energy and thermal energy are the driving forces for the recrystallization process in the cold-rolled HSLA-V steels. It is also notable that our fitted t_s (time by when nucleation stops) decreases with an increase in the cold-

rolling reduction and annealing temperature. Thus, our model analysis revealed that the recrystallization process in the HSLA-V steels processed with high cold-rolling reduction and high annealing temperature is mainly the growth of recrystallized grains with high kinetic rate whereas in the HSLA-V steels processed with low cold-rolling reduction and low annealing temperature consists of distinct nucleation stage and grain growth stage.

Figure 8. Measured (circles) and modelled (curves) recrystallized volume fraction varies as a function of annealing time in the HSLA-V steels which had been cold-rolled by (a) 40%, (b) 60%, and (c) 80 % reduction and subsequently annealed at various temperatures (i.e., 630 °C, 650 °C, and 670 °C).

Table 2. Processing and kinetic parameters of the recrystallized HSLA-V steels in this study. The recrystallization kinetic parameters of the HSLA-V steels were obtained by fitting our analysis model (Eq. (4)) to the experimental data of recrystallization rate (shown in Fig. 8) under the assumption that the recrystallization growth dimension m is 1.

Processing Parameters		Kinetic Parameters	
Cooled-rolling Reduction (%)	Annealing Temperature (°C)	Recrystallization Rate C ($\times 10^{-8}s^{-3/2}$)	Nucleation Stop Time t_s (hr)
40	630	5.58	8.06
40	650	48.8	1.11
60	630	169	1.25
60	650	604	0.39
60	670	1396	0.19
80	630	1036	0.28
80	650	8040	0.06

3.4. Recrystallization kinetics extracted from model analysis

Furthermore, we extracted the recrystallization kinetics of the HSLA-V steels using the data in Table 2.

Considering the recrystallization process (i.e., the nucleation and grain growth) to be a thermally activated process, we can express the kinetic rate C as a function of annealing temperature in an Arrhenius relationship.

$$C = C_T \exp(-Q_C/RT) \quad (5)$$

where, Q_C is the activation energy for recrystallization and R is the gas constant. It should

be pointed out that the total activation energy Q_C for recrystallization includes both the activation energy for grain nucleation, which is the energy associated with new interfacial surface generation, and the activation energy for grain growth, which is the energy associated with grain boundary migration [35].

Our results in Table 2 indicated that an increase in the cold-rolling reduction would also lead to an increase in the recrystallization kinetic rate C of the HSLA-V steels. This is because that the high cold-rolling reduction deformation would produce an increment in the dislocation density in the sample and the high-density dislocations provide enhanced sources for nucleation and driving force to sustain grain growth [44]. Taking the strain effect into account, we can further express the kinetic rate C as a function of both annealing temperature and cold-rolling reduction (ϵ) as follows.

$$C = C_0 \epsilon^{q_c} \exp(-Q_C/RT) \quad (6)$$

Here, we assume that the parameters C_0 , Q_C and q_c are constant independent of the cold-rolling reduction and annealing temperature. By fitting the kinetic rate C against the two processing parameters in Table 2, we determined rate C_0 to be $1.65 \times 10^{23} \text{ s}^{-\frac{3}{2}}$, activation energy Q_C to be 474 kJ/mol, and the exponent q_c on strain to be 7.419. In comparison, Mukunthan et al [45] found that the recrystallization kinetics in an 80% cold-rolled Ti-Nb low carbon steel exhibited an apparent JMAK exponent of 0.73 and activation energy of 501.7 kJ/mol in the isothermal annealing temperature range between 500 to 760°C. In addition, they observed that the nucleation of recrystallized grains occurred primarily during the early stages of recrystallization process, in consistent with our kinetic analysis model. Considering the similarity in the steels and experimental conditions, we believe that the agreement between our work and Ref. [45] indicates that our recrystallization kinetic analysis model could be applied to a broad range of microalloyed low carbon steels.

As discussed above, our physics-based model in Eq. (4) captures excellently the

recrystallization kinetics in the annealed cold-rolled HSLA-V steels with some selected processing parameters. Furthermore, we can extend our model to produce predictions for the recrystallized HSLA-V steels processed at conditions which were not tested in the experiments and thus enable a processing design for the HSLA-V steels. For instance, if some engineering application requires a 90% recrystallized fraction to eliminate the residual stress in the steel, our model predicts that it would take 6 hour annealing at 650 °C for the HSLA-V steels with a 80% cold-rolling reduction to achieve that value, but need 21 hour annealing at 650 °C for the HSLA-V steels with a 60% cold-rolling reduction to achieve the same goal. It should be mentioned that our analysis model still requires a small amount of experimental tests to determine its parameters, such as tunable parameter t_s , for recrystallization kinetics prediction.

4. Conclusions

In this work, we studied the annealing process in the cold-rolled Vanadium microalloyed HSLA steels using both the experimental characterization for microstructure evolution and the theoretical analysis for recrystallization kinetics. The following conclusions are drawn from our study.

- (1) The full-hard cold-rolled HSLA-V steel has a microstructure consisting of elongated grains along the rolling direction and a high stored energy associated with high density of deformation dislocations. During a subsequent annealing, the stored energy in the cold-rolled HSLA steel is released through the nucleation and growth of equiaxed recrystallization grains in replacement of the deformed grains.
- (2) From experimental characterization study, we attained a dataset of the stored energy and volume fraction of recrystallization as a function of annealing time in the HSLA-V steel samples which have been cold-rolled by 40 %, 60 % and 80 % reduction and further annealed at 630 °C, 650 °C, and 670 °C, respectively. The recrystallization kinetics in the HSLA-V steels is strongly dependent on the processing parameters of

cold-rolling reduction extent and annealing temperature.

- (3) A modified JMAK model was developed to describe the recrystallization kinetics in the cold-rolled HSLA-V steels. The predictions from our model agree excellently with the recrystallization kinetics measured from the experiments.
- (4) Our study reveals that the recrystallization process in the cold-rolled HSLA-V steels follows one dimensional growth of the recrystallized grains and requires thermal activation energy of about 474 kJ/mol.

Acknowledgment

G.F. Wang would like to acknowledge the research grant National Science Foundation (Grant No. DMR-1410597). The authors gratefully acknowledge the computational resources provided by the computer facility at Center for Simulation and Modeling of the University of Pittsburgh and at the Extreme Science and Engineering Discovery Environment (XSEDE), which is supported by National Science Foundation grant number ACI-1053575.

References

- [1] G. Murray, Handbook of Materials Selection for Engineering Applications, CRC Press, Boca Raton, 1997.
- [2] M.S. Rashid, Strain-aging of vanadium, niobium or titanium-strengthened high-strength low-alloy steels, Metall. Mater. Trans. A 6 (1975) 1265-1268.
- [3] M.S. Rashid, High-strength, low-alloy steels, Science 208 (1980) 862-869.
- [4] R. Steiner, ASM Handbook Volume 1: Properties and Selection: Irons, Steels, and High-Performance Alloys, ASM International, Materials Park, 1990.
- [5] J. Majta, K. Muszka, Mechanical properties of ultra fine-grained HSLA and Ti-IF steels, Mater. Sci. Eng. A 464 (2007) 186-191.
- [6] A. Galibois, M. R. Krishnadev, A. Dubé, Control of grain size and sub-structure in plain carbon and high strength low alloy (HSLA) steels—the problem and the prospect, Metall. Mater. Trans. A 10 (1979) 985-995.
- [7] M.-Y. Chen, M. Gouné, M. Verdier, Y. Bréchet, J.-R. Yang, Interphase precipitation in vanadium-alloyed steels: strengthening contribution and morphological variability with austenite to ferrite transformation, Acta Mater. 64 (2014) 78-92.
- [8] M. A. Altuna, Amaia Iza-Mendia, I. Gutiérrez, Precipitation of Nb in ferrite after austenite conditioning. Part II: strengthening contribution in high-strength low-alloy (HSLA) Steels, Metall. Mater. Trans. A 43 (2012) 4571-4586.
- [9] L. Luyckx, J. R. Bell, A. McLean, M. Korchynsky, Sulfide shape control in high strength low alloy steels, Metall. Trans. 1 (1970) 3341-3350.
- [10] S. K. Das, S. Chatterjee, S. Tarafder, Effect of microstructures on deformation behaviour of high-strength low-alloy steel, J. Mater. Sci. 44 (2009) 1094-1100.
- [11] T. N. Baker, Processes, microstructure and properties of vanadium microalloyed steels, Mater. Sci. Tech. 25 (2009) 1083-1107.

- [12] B. K. Show, R. Veerababu, R. Balamuralikrishnan, G. Malakondaiah, Effect of vanadium and titanium modification on the microstructure and mechanical properties of a microalloyed HSLA steel, *Mater. Sci. Eng. A* 527 (2010) 1595-1604.
- [13] R. Lagneborg, T. Siwecki, S. Zajac, B. Hutchinson, The role of vanadium in microalloyed steels, *Scand. J. Metall.* 28 (1999) 186-241.
- [14] M. Janošec, I. Schindler, J. Palát, L. Čížek, V. Vodárek, E. Místecký, M. Růžička, L.A. Dobrzański, S. Rusz, P. Suchánek, Properties of a Nb-V-Ti microalloyed steel influenced by cold rolling and annealing, *J. Achievements Mater. Manuf. Eng.* 20 (2007) 251-254.
- [15] R. Song, D. Ponge, D. Raabe, J.G. Speer, D.K. Matlock, Overview of processing, microstructure and mechanical properties of ultrafine grained bcc steels, *Mater. Sci. Eng. A* 441 (2006) 1-17.
- [16] N. Prasad, S. Kumar, P. Kumar, S. N. Ojha, Mechanical properties of a cold-rolled annealed HSLA steel, *J. Mater. Sci.* 26 (1991) 5158-5162.
- [17] G. F. Vander Voort, *Metallography, Principles and Practice*, ASM International, Materials Park, 1984.
- [18] M. Avrami, Kinetics of phase change. I general theory, *J. Chem. Phys.* 7 (1939) 1103-1112.
- [19] M. Avrami, Kinetics of phase change. II transformation - time relations for random distribution of nuclei, *J. Chem. Phys.* 8 (1940) 212-224.
- [20] W. A. Johnson, R. F. Mehl, Reaction kinetics in processes of nucleation and growth, *Trans. AIME* 135 (1939) 416-458.
- [21] A. Kolmogoroff, Zur statistik der kristallisationsvorgänge in metallen, *Izv. Akad. Nauk Ser. Mater.* 1 (1937) 355-359.
- [22] S. H. Choi, Y. S. Jin, Evaluation of stored energy in cold-rolled steels from EBSD data, *Mater. Sci. Eng. A* 371 (2004) 149-159.

- [23] D. A. Porter, K. E. Easterling, M. Y. Sherif, Phase Transformations in Metals and Alloys, 3rd ed., CRC Press, Boca Raton, 2009.
- [24] S. Dzaszyk, E.J. Payton, F. Friedel, V. Marx, G. Eggeler, On the characterization of recrystallized fraction using electron backscatter diffraction: A direct comparison to local hardness in an IF steel using nanoindentation, *Mater. Sci. Eng. A* 527 (2010) 7854–7864.
- [25] N. Peranio, Y. J. Li, F. Roters, D. Raabe, Microstructure and texture evolution in dual-phase steels: competition between recovery, recrystallization, and phase transformation, *Mater. Sci. Eng. A* 527 (2010) 4161-4168.
- [26] S. Mandal, S. K. Mishra, A. Kumar, I. Samajdar, P. V. Sivaprasad, T. Jayakumar, Baldev Raj, Evolution and characterization of dynamically recrystallized microstructure in a titanium-modified austenitic stainless steel using ultrasonic and EBSD techniques, *Philos. Mag.* 88 (2008) 883-897.
- [27] S. Zaefferer, P. Romano, F. Friedel, EBSD as a tool to identify and quantify bainite and ferrite in low - alloyed Al - TRIP steels, *J. Microsc.* 230 (2008) 499-508.
- [28] R. A. Vandermeer, P. Gordon, in *Recovery and Recrystallization of Metals*, Himmel, L. ed., Interscience, New York, 1963, 211.
- [29] I. Samajdar, B. Verlinden, P Van Houtte, D. Vanderschueren, Recrystallisation kinetics in IF-steel: a study on the sluggish recrystallisation behaviour, *Scripta Mater.* 37 (1997) 869-874.
- [30] R. A. Vandermeer, D. J. Jensen, The migration of high angle grain boundaries during recrystallization, *Interface Sci.* 6 (1998) 95-104.
- [31] G. Wu, D. Juul Jensen, Recrystallization kinetics of aluminum AA1200 cold rolled to true strain of 2. *Mater. Sci. Tech.* 21 (2005) 1407-1411.
- [32] C.M. Sellars, Effect of varying hot deformation conditions on deformed microstructures and subsequent static recrystallization, in: T.R. McNelly (Ed.),

Proceedings of the 3rd Conference on Recrystallization and Related Phenomena, 1997, pp. 81–93.

[33] H. Luo, J. Sietsma, S. van der Zwaag, Characteristics of the static recrystallization kinetics of an intercritically deformed C-Mn steel, *Mater. Sci. Forum* 467-470 (2004) 293-298

[34] M. Oyarzábal, A. Martínez-de-Guerenu, I. Gutiérrez, Effect of stored energy and recovery on the overall recrystallization kinetics of a cold rolled low carbon steel. *Mater. Sci. Eng. A* 485 (2008) 200-209.

[35] F. J. Humphreys, M. Hatherly, *Recrystallization and Related Annealing Phenomena*, Elsevier, 2nd ed., Oxford, 2004.

[36] Z. Li, Z. Yang, C. Zhang, Z. Liu, Influence of austenite deformation on ferrite growth in a Fe–C–Mn alloy, *Mater. Sci. Eng. A* 527 (2010) 4406-4411.

[37] R. D. Doherty, D. A. Hughes, F. J. Humphreys, J. J. Jonas, D. Juul Jensen, M. E. Kassner, , W. E. King, T. R. McNelley, H. J. McQueen, A. D. Rollett, Current issues in recrystallization: a review, *Mater. Sci. Eng. A* 238 (1997) 219-274.

[38] C. W. Price, Use of Kolmogorov-Johnson-Mehl-Avrami kinetics in recrystallization of metals and crystallization of metallic glasses, *Acta Metall. Mater.* 38 (1990) 727-738.

[39] C. García-Mateo, B. Lopez, J. M. Rodríguez-Ibabe, Static recrystallization kinetics in warm worked vanadium microalloyed steels *Mater. Sci. Eng. A* 303 (2001) 216-225.

[40] R. Thibau, J. Masounave, L. Piperni, Recrystallization model for controlling mechanical properties of batch-annealed cold-rolled low-carbon aluminum-killed steel sheets, *Mater. Sci. Tech.* 2 (1986) 1038-1045.

[41] Z. Wang, X. Sun, Z. Yang, Q. Yong, C. Zhang, Z. Li, Y. Weng, Carbide precipitation in austenite of a Ti–Mo-containing low-carbon steel during stress relaxation, *Mater. Sci. Eng. A* 573 (2013) 84-91.

- [42] S. Shanmugam, M. Tanniru, R. D. K. Misra, D. Panda and S. Jansto, Microalloyed V–Nb–Ti and V steels Part 2–Precipitation behaviour during processing of structural beams, *Mater. Sci. Tech.* 21 (2005) 165-177.
- [43] C. P. Reip, S. Shanmugam, R. D. K. Misra, High strength microalloyed CMn (V–Nb–Ti) and CMn (V–Nb) pipeline steels processed through CSP thin-slab technology: Microstructure, precipitation and mechanical properties, *Mater. Sci. Eng. A* 424 (2006) 307-317.
- [44] C. Lingk, M. E. Gross, Recrystallization kinetics of electroplated Cu in damascene trenches at room temperature, *J. Appl. Phys.* 84 (1998) 5547-5553.
- [45] K. Mukunthan, E. B. Hawbolt, Modeling recovery and recrystallization kinetics in cold-rolled Ti-Nb stabilized interstitial-free steel, *Metall. Mater. Trans. A* 27 (1996) 3410-3423.