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THE EVOLUTION OF AUSTENITE RECRYSTALLIZATION AND GRAIN GROWTH USING LASER-ULTRASONICS

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ABSTRACT

Laser-ultrasonics is a non-contacting technique whereby the attenuation of ultrasonic signals can be measured and related to the grain size of the investigated material. In the present paper, a laser-ultrasonic grain size measurement technique previously developed for various C-Mn and microalloyed steels has been extended to examine austenite recrystallization and subsequent grain growth following hot deformation. The ultrasonic measurements were conducted on a low carbon (0.05wt%) steel that contains Mn, Mo and Nb as the three main microalloying elements. The grain size data measured by ultrasonic experiments was analyzed to quantify the effect of deformation conditions on the evolution of recrystallized grain size and subsequent grain growth. A significant effect of deformation temperature, applied strain and initial grain size on grain size evolution was observed while strain rate had a negligible effect. Phenomenological modeling approaches were employed to describe recrystallized grain size and grain growth behaviour of the present steel.

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I. INTRODUCTION

Austenite grain size is an important microstructural parameter during hot rolling of steels [1-3]. The austenite grain size influences the softening behaviour during rolling and the subsequent phase transformation during cooling and therefore affects the final microstructure and mechanical properties. In modern microalloyed low carbon steels, a careful choice of thermomechanical processing route and various grain refiners such as niobium (Nb), titanium (Ti), or molybdenum (Mo) can be employed to achieve the desired final microstructures (i.e. after austenite decomposition) [1]. Generally, these grain refiners control the austenite grain structure by i) retarding austenite recrystallization and grain growth and ii) preserving pancaked austenite at the finish mill exit by particle pinning or solute drag [4-8]. In particular, Nb is very effective in delaying recrystallization [9-12]. Given the significance of the austenite microstructure, it is important to have reliable knowledge of the austenite grain size evolution during reheating and hot rolling.

From an experimental perspective, the investigation of the austenite microstructure is challenging, as this microstructure is not present at room temperature. However, over the years, a number of metallographic tools have been developed to measure austenite grain structures. These tools include conventional metallography, thermal grooving, oxidation, carburization, glass etching and ferrite/cementite delineation of prior austenite grain boundaries [13]. All these techniques have their advantages and limitations. Conventional metallography involves heat treating the sample in the austenite temperature range followed by quenching. Provided a martensite microstructure results from the quench, suitable etching procedure can be employed to reveal the prior austenite grain boundaries.

However, for low carbon steels with low hardenability, excessive quench rate (>1000K/s) may be required to produce martensite and alternative techniques have to be employed. Some other techniques, e.g. thermal grooving, glass etching, oxidation or carburization of austenite grain boundaries are limited to surface grains where the grain structure may be different from that of the bulk [13, 14]. In addition, none of the above techniques can be employed for real-time monitoring of austenite grain size at high temperatures.

Laser-ultrasonics is an attractive alternative for real-time monitoring of grain size evolution in austenite. Ultrasonic attenuation, i.e. the decay of an ultrasonic wave as it propagates through the material, is sensitive to material parameters such as grain size [15, 16] and porosity [17]. Thus, in the absence of porosity, attenuation data can be directly correlated to the austenite grain size [14, 16, 18, 19]. Further, the change in relative velocity of the ultrasound wave during its propagation through the material is related to the evolution of texture and can be utilized to monitor recrystallization in-situ [16, 20-22]. Laser-ultrasonics is a remote, continuous and non-destructive technique that can be operated online at high temperatures for bulk observation [16, 18, 23-25].

Laser-ultrasonic measurements of the austenite grain size have so far emphasized conventional grain growth tests that involve reheating of a sheet sample [14, 18, 19, 26]. Based on these investigations, calibration procedures were developed that relate the measured attenuation to the austenite grain size for plain carbon and microalloyed steels with carbon contents ranging from 0.05 to 0.74wt% [14, 18]. Further, initial laser-ultrasonic measurements were made in combination with stress relaxation technique to monitor in-situ recrystallization and recrystallized grain size for a C-Mn steel [22].

The present paper describes laser-ultrasonic measurements of austenite grain size evolution after deformation; i.e. a problem relevant to commercial hot rolling of steel. For this purpose modified measurement procedures were established to obtain information on recrystallization, recrystallized grain size and austenite grain growth. The tests were conducted on a 0.05wt% carbon steel where conventional austenite grain growth had previously been studied by laser-ultrasonics [26]. This approach offers insight into austenite grain growth after recrystallization that is considered in the framework of phenomenological microstructure evolution models.

II. EXPERIMENTAL

The chemistry of the low-carbon steel examined is shown in Table I. The steel was laboratory cast as a model material for a complex-phase steel and received in the form of forged bars. Cylindrical samples (15mm in length and 10mm diameter) were machined for axisymmetric compression tests using a Gleeble 3500 thermo-mechanical simulator equipped with a laser-ultrasonic system. The surfaces of the samples were machined to a smooth finish to minimize the effect of surface roughness on ultrasound attenuation. During the tests, a vacuum of approximately 7Pa (50millitorr) was maintained in the Gleeble chamber. A thermocouple was spot-welded to the center of the sample; a K type thermocouple was employed for soaking the sample at 1000°C, whereas a R type thermocouple was used for higher soaking temperatures. Temperature gradients were evaluated with a second thermocouple placed near the end of the sample for some initial tests and were found to be in the order of 5°C. To reduce friction, graphite foil in combination with nickel paste was used as lubricant. The test matrix is shown in Table II.

The initial soaking conditions were selected to achieve a uniform austenite grain structure. Significant abnormal grain growth had been observed for the present steel during soaking at temperatures between 1100-1200°C [26]. Thus, these temperatures were avoided for the selection of initial soaking conditions. A heating rate of 5°C/s was employed to heat the sample up to the soaking temperature. The sample was soaked there for 120s followed by cooling at 10°C/s to the deformation temperature. After holding for 5s at the designated deformation temperature, the sample was compressed with applied strain and strain rates, as indicated in Table II. After deformation, the sample was held at the deformation temperature for 10min before being rapidly cooled by He quench to room temperature. The fast cooling step was important to acquire reference ultrasonic data at room temperature.

The ultrasonic measurements were taken from the beginning of deformation and continued till the end of the experiment at room temperature. The laser generating and detecting spots were set-up in such a way that after deformation, the thermocouple, generating laser and detecting laser remained on the same vertical plane. A schematic diagram of the experimental set-up is shown in Figure 1. To generate the ultrasonic wave, a short (5ns), energetic (150mJ) green (532nm) light pulse from a frequency-doubled Nd:YAG laser was employed. This light pulse generates a wideband ultrasound pulse by ablating a thin surface layer. The laser interferometer employs a longer pulse Nd:YAG laser operating at 1064nm (infrared), with a pulse duration of 50μs and pulse energy of approximately 70mJ. The detection laser beam was focused on the sample into a uniform disk approximately 2mm in diameter, i.e. roughly the same diameter as the source laser spot that was carefully aligned prior to the test on the opposite side of the cylindrical

sample by monitoring the propagation delay of the ultrasound pulse. A confocal Fabry-Perot interferometer operating in reflection and with a detection bandwidth of 75MHz was employed. The acquisition rate was initially set to 10Hz to capture the rapid microstructure variations following the deformation, and gradually lowered to about 0.2Hz during the experiment to avoid any possible depression mark on the sample which could have occurred if an excessively large number of measurements had been taken. Although, ultrasonic measurements were carried out during and after deformation the grain size evolution analysis was performed when the piston stopped moving forward and the first time point was defined to occur as t=0.05s (0.05s is half the measurement period). A smoothing function (10 point adjacent average method) was used to represent the experimental data.

III. LASER-ULTRASONICS: METHODS AND PROCEDURES

A. *Measurement Principle*

To measure the austenite grain size on cylindrical samples after hot deformation, a new laser-ultrasonic measurement procedure was developed. To adequately explain the new measurements, it is necessary to review the principles of grain size measurements by laser-ultrasonics. Ultrasonic grain size measurements are based on the grain size dependence of ultrasonic attenuation [14, 15, 27]. This may be written as:

$$\alpha_{Sc}(f,T) = K(T)D^{n-1}f^{n} \qquad [1]$$

where α_{Sc} is the ultrasonic attenuation resulting from scattering by the grains, f is the frequency of the ultrasound wave, T is the operating temperature, D is the average grain diameter, and $K(T)$ is a parameter that depends on temperature. It is known from

theoretical considerations that $0 \leq n \leq 4$ depending on the ratio of acoustic wavelength to grain diameter. In the limit of wavelengths much larger than the grain diameter, n tends towards 4 whereas in the limit of wavelengths much smaller than the grain diameter n tends towards 0. In practice, at very long wavelengths, the total attenuation is small and difficult to measure. Conversely, at very short wavelength, the attenuation is so strong that the ultrasound is completely scattered by a few grains. Even though n varies in principle with frequency, in practice, it can be assumed constant (usually n is in the range of 1.5 to 3.5), even when a relatively large frequency range is employed as shown for stainless steel [28].

In addition, other important phenomena such as geometrical considerations, the associated self-diffraction of ultrasound, and internal friction [29] can affect signal amplitude and thus contribute to the total attenuation. When an ultrasound pulse travels some distance along the x direction, from $x=x_1$ to $x=x_2$, the total attenuation may be expressed as:

$$\alpha(x_1, x_2, f, T) = \frac{20}{x_2 - x_1} \log \frac{A(x_1, f, T)}{A(x_2, f, T)} = \alpha_{IF}(f, T) + \alpha_{Sc}(f, T) + \alpha_D(x_1, x_2, f) \qquad [2]$$

where, α_{IF} and α_D are the contributions to attenuation from internal friction and diffraction respectively and $A(x, f, T)$ is the amplitude of a pulse that has traveled a distance x. In the previous work [14], the geometry was chosen such that the acoustic wave was a plane wave so that $\alpha_D=0$. In addition, the grain sizes were relatively large compared to the wavelength of the ultrasound wave and n was found to be 1.5. This means that scattering by the grains was very intense such that the contribution of internal

friction could be neglected and the total attenuation was due to scattering. i.e. $\alpha = \alpha_{Sc}$ as given in Eq. 1.

An alternative measurement can be used when the geometry cannot be chosen such that α_D is zero or when it is not known precisely. In such a case, α_D can be estimated experimentally by using another sample having the same sound velocity and geometry and having zero (or negligible) internal friction and scattering. In previous work [18, 26], this reference sample was the same sample or a sample of the same shape and material, but with the measurements taken at room temperature, prior to heat treatment. Using the subscript "*ref*" to identify measurements made on the reference sample one has

$$\alpha_{ref}(x_1, x_2, f, T) = \frac{20}{x_2 - x_1} \log \frac{A(x_1, f, T_{ref})}{A(x_2, f, T_{ref})} = \alpha_D(x_1, x_2), \qquad [3]$$

and Eq. 2 becomes

$$\frac{20}{x_2 - x_1} \log \left(\frac{A(x_1, f, T)}{A(x_2, f, T)} \frac{A(x_2, f, T_{ref})}{A(x_1, f, T_{ref})} \right) = \alpha_{IF}(f, T) + \alpha_{Sc}(f, T) \qquad [4]$$

In general, the measurement sensitivity of laser-ultrasonics varies with temperature and surface condition of the sample as they affect the amplitude of the generated ultrasound pulse and the sensitivity of the detection interferometer. However for the ultrasound wave that has not yet propagated within the sample (i.e. the wave at $x=0$) it can be assumed that the ultrasound pulse amplitude, and not its shape, depends on temperature. This is a reasonable assumption if the reference sample is made of the same material as the materials to be tested. Therefore, the pulse amplitude at one temperature can be linearly related to the pulse amplitude at another temperature, i.e.

$$A(0, f, T) = c(T)A(0, f, T_{ref}) \qquad [5]$$

where c is a parameter that can vary with temperature and the sample condition. Implicit in Eq.5 is the assumption that the frequency dependence (pulse shape) of the generated ultrasound pulse is independent of temperature. Letting $x_1=0$ and $x_2=x$ in Eq.4, one has

$$\frac{20}{x}\log\left(\frac{A(x,f,T_{ref})}{A(x,f,T)}\right) = -\frac{20\log c}{x} + \alpha_{IF}(f,T) + \alpha_{Sc}(f,T) \qquad [6]$$

Eq.6 shows that only two measurements are needed: one at temperature of interest and the other at a reference temperature for the same propagation distance. As an improvement to the method of Dubois et al. [14] where internal friction was assumed to be zero, it can be assumed that internal friction contributes an unknown but frequency-independent amount to the total attenuation. Eq.6 combined with Eq.1 becomes

$$\frac{20}{x}\log\left(\frac{A(x,f,T_{ref})}{A(x,f,T)}\right) = a + \alpha_{Sc}(f,T) = a + bf^n \qquad [7]$$

where $a = -\dfrac{20\log c}{x} + \alpha_{IF}(T)$ and b is related to grain size by

$$b = K(T)D^{n-1} \qquad [8]$$

In Eq.7, the left side is measured experimentally. Then, it is fit to the right side of Eq.7 where, a, b and n are the fitting parameters. Although there are models to estimate $K(T)$ and n, it is more practical to obtain them empirically. This amounts to a calibration procedure where b and D are measured simultaneously and n is assumed constant for some set of calibration samples. Such a calibration is available for austenite grain size in C-Mn and microalloyed steels where $n = 3$ was assumed [18]. In this previous work, the reference sample was chosen as a steel sample of low attenuation having the same dimensions, or as the same steel sample prior to being heated to high temperature (provided the scattering is sufficiently low prior to heating). However the above

procedure is not acceptable in the present work due to the dimensional changes caused by the deformation at high temperature. Thus, a suitable reference sample is obtained at the end of the experiment by rapidly cooling to room temperature to attain a fine-scale microstructure that ensures much lower scattering at room temperature than at high temperature.

B. *Measurement Geometry*

Prior measurements of austenitic grain size were done either on sheet and plate samples [14, 26] or on tubes [18]. The ultrasound pulses were simple pressure pulses (longitudinal waves), reflected back and forth between the two free surfaces. However the present study employs samples of cylindrical geometry that complicate measurements. When ultrasound is excited with a localized pulsed excitation, multiple acoustic waves propagate: a pressure pulse across the diameter of the cylinder, a surface wave around the circumference of the cylinder, and pressure pulses traveling in various directions that are reflected and mode converted to shear pulses by the curved surfaces of the cylinder [30, 31]. This can lead to multiple pulses that are difficult to identify, or worse, that are partly superimposed.

The simplest possible geometry and propagation path was to locate the excitation and detection of the acoustic pulses on the opposite sides, across the diameter of the cylinder. The acoustic pulse to be measured is simply the first pulse to travel across the diameter. This pressure pulse is easy to identify because it has a high amplitude, the highest velocity (as compared to other waves), and the shortest path. In addition, the present work involves small enough generation and detection spots of the ultrasound (an area of

less than 2mm diameter) so that they nearly behave as point-like transducers having smooth and well-behaved ultrasound radiation patterns.

C. *Calibration*

A previous study [26] established that a developed calibration procedure based on the C-Mn and microalloyed steels [18] can be employed successfully to monitor austenite grain growth for the current steel using a simple rectangular strip geometry. Figure 2 shows the validation results at an austenite temperature of 1050°C for the present scenario, i.e. where the sample has a cylindrical geometry. The metallographic data were taken from the previous study [26]. The present ultrasonic results are in good agreement with metallographic data confirming that the employed experimental procedure accounts adequately for the sample geometry. Although the ultrasonic measurements show some scatter, the grain size evolution as measured by laser-ultrasonics shows reproducibility within the error margins of approximately 20-25%.

IV. ULTRASONIC RESULTS: EXPERIMENTAL AND MODELLING

A. *General Observations*

With the previously developed calibration procedure being validated, the measured attenuation data for the deformation tests can be represented in terms of the austenite grain size. Figure 3 presents the ultrasonic grain size evolution for various conditions in terms of temperature, deformation strain, initial grain size, and strain rate. The results of the individual measurements show three distinct stages:

grain size values (from 272μm to 152μm). An average grain size value of 205μm is determined for reheating at 1250°C. For each of the other reheating temperatures, i.e. for 1000, 1050 and 1100°C, the present study has only one experimental grain size value (Table II). Thus for the latter reheating conditions, the measured austenite grain size values are compared with the previously determined austenite grain sizes by laser-ultrasonics for the same steel grade [26]. Both results agree with each other within the experimental error of approximately 20-25%. Thus, initial austenite grain size values as measured here, i.e. 52, 17 and 14μm for soaking temperatures of 1100, 1050, and 1000°C, respectively, are adopted for further analysis.

C. *Recrystallized Grain Size*

Figure 4 shows the recrystallized grain size values obtained from ultrasonic measurements. For a given deformation temperature (1000°C), the recrystallized grain size decreases with applied strain and increases with initial austenite grain size. Further, there is no effect of strain rate on recrystallized grain size. These trends are consistent with those reported previously based on metallographic analysis [12]. The situation is more complex when the variation with deformation temperature is considered. As shown in Figure 4(a), the recrystallized grain size exhibits minimal changes when the deformation temperature is increased from 1000°C to 1100°C, whereas the grain size increases significantly at higher deformation temperature (1150°C). Previous study suggested significant grain growth for an operating temperature of 1150°C and above [26]. Thus, one possible explanation for the significantly higher recrystallized grain size values at 1150°C is that recrystallization and substantial grain growth may overlap at

these higher temperatures resulting in a significantly larger recrystallized grain size value when the material achieves the fully recrystallized state. This complex behaviour of the temperature dependence of the recrystallized grain size reflects the variety of relationships proposed in the literature that show either a temperature-independent or a weakly temperature-dependent recrystallized grain size [12, 33].

Frequently, an empirical relationship is employed to describe the recrystallized grain size ($D_{Rx.}$) as a function of initial grain size (D_0), applied strain (ε) and deformation temperature (T) [7, 12, 33]:

$$D_{Rx.} = CD_0^p \varepsilon^q \exp\left(-\frac{Q_{gx.}}{RT}\right)$$ [9]

where C, p, q and $Q_{gx.}$ are adjustable parameters. In the present analysis $Q_{gx.}=0$ (i.e. the recrystallized grain size is independent of temperature) for T≤1100°C and $Q_{gx.}=165$kJ/mol for T>1100°C are adopted together with $C=0.35\mu m^{1-p}$ for T≤1100°C and $C=6\times10^5$ μm^{1-p} for T>1100°C. In addition, $p=0.84$, $q=-0.83$ are determined from the present analysis. For comparison, the model descriptions are included in Figure 4 and show satisfactory agreement with the experimental observation.

D. *Grain growth*

Figure 5 shows the grain growth behaviour after recrystallization when the sample was held at the deformation temperature for approximately 10min. Little grain growth is observed at lower temperatures, such as 1000°C and 1050°C (91μm to 140μm at 1000°C and 80μm to 128μm at 1050°C within a time span of 10min). When the temperature is increased to 1150°C, substantial grain growth occurs, i.e. the grains grow from 128μm to

285μm within 5min. These trends are consistent with the austenite grain growth observations made in the previous laser-ultrasonic study of the present steel [26]. The comparatively limited grain growth at lower temperatures is probably due to particle pinning and/or solute drag effects whereas these effects are much weaker at higher temperatures. Further, as observed, the applied strain and strain rate have little effect on subsequent grain growth. However, the present conditions are quite different from those of conventional grain growth tests that only involve a simple reheating step. To appreciate the observed grain growth kinetics, a more rigorous analysis of the data was performed using established model concepts for grain growth.

Burke and Turnbull [34, 35] developed the original grain growth model by assuming that the pressure on the boundary arises from the curvature of the boundary. This leads to a parabolic grain growth law. However, many observations of grain growth, even in ultra-pure materials, suggest deviations from the parabolic growth behaviour such that frequently an empirical power law is proposed for grain growth:

$$D^m = D_0^m + Kt \qquad\qquad [10]$$

where, D_0 is the initial grain size, K is a temperature-dependent material parameter, t denotes time, and m is the grain growth exponent that is usually larger than 2. The present grain growth data suggests grain growth exponents in the range of 3.5-5. These deviations from the ideal parabolic grain growth behaviour have been attributed to the effects from solute drag and particle pinning [35]. Both aspects are important for the present steel as it contains Nb and Mo, which are strong carbide formers and, in addition, exhibit significant solute drag effects [7, 9-12]. De-convolution of both mechanisms is challenging. Solute drag is usually described with a reduced effective mobility that,

Initially, the grain size remains almost constant for some time just after deformation (varying from approximately 1s to 10s depending on the experimental condition). This suggests that this initial stage represents the time interval before the onset of recrystallization where recovery is the dominant softening process. The times for 5% recrystallization as predicted by an independently developed softening model [32] are indicated by open symbols in Figure 3 and agree well with the end of the initial regime as measured by laser-ultrasonics.

The second regime exhibits a marked decrease of grain size and coincides with recrystallization during this period. New grains begin to appear and in the process the average grain size becomes finer. The time for 95% recrystallization predicted by the softening model [32] is indicated in Figure 3 by open symbols. These times are in agreement with the position of the minimum grain size measured by laser-ultrasonics at the end of this second period for 9 out of 10 deformation conditions as shown in Figure 3 (the one exception is for an applied strain rate of 0.1/s, see Figure 3c). Thus, the minimum grain size will be considered as the recrystallized grain size. In the final stage, the grain size increases for all deformation situations. This observation is consistent with grain growth after the completion of recrystallization.

B. *Initial Grain Size*

The initial grain size for each reheating condition was evaluated by averaging the grain size values measured by ultrasonics during the first regime; the results are shown in Table II as a function of reheating condition. As can be observed from Table II, for reheating temperature of 1250°C, there is a significant variation in the initial austenite

however, would not lead to deviations from $m=2$. To account for $m>2$ due to solute drag, a transition from a high to a low velocity branch during grain growth would have to be considered. In contrast, introducing a pinning factor is a well established procedure to explain $m>2$ [35]. Thus, here an approach for grain growth modelling that includes a pinning term (P) is selected. The generalized grain growth equation is then:

$$\frac{d\overline{D}}{dt} = M_{GB}\left(\frac{\alpha\gamma_{GB}}{\overline{D}} - P\right) \qquad [11]$$

where, $\overline{D}$ is the average austenite grain size, M_{GB} is the effective grain boundary mobility, α is a geometrical constant and γ_{gb} is the grain boundary energy. For the present calculations, $\alpha=0.5$ and a grain boundary energy value of $0.75J/m^2$ are used [34, 35]. The grain boundary mobility is expressed as:

$$M_{GB} = M_0 \exp\left(\frac{-Q_{eff.}}{RT}\right) \qquad [12]$$

where, M_0 is the pre-exponential factor, $Q_{eff.}$ is an effective grain boundary activation energy, and R is the universal gas constant $(8.3145Jmol^{-1}K^{-1})$. In the present analysis, $Q_{eff}=330kJ/mol$ is adopted from a previous softening study on the present steel [32]. Then, the pinning term P and M_0 are employed as fitting parameters. Here, M_0 is a constant whereas P may depend on process conditions (e.g. temperature). Using this approach, $M_0=1.9\times10^9m^4/Js$ is determined and the values obtained for P are listed in Table III. Figure 5 compares the model description for grain growth with the experimental data. The present model reflects the grain growth behaviour reasonably well when M_0 is fixed and P is considered to depend on each grain growth situation.

V. DISCUSSION

A new laser-ultrasonic technique was developed in the present research to measure in-situ the austenite grain size evolution after deformation in a low carbon steel. The present method enables to apply laser-ultrasonics as a grain size measurement technique when the sample geometry is complex and/or changed due to plastic deformation. The accuracy of these measurements is similar to those made previously using simpler specimen geometries, i.e. tube or sheet [18, 26]. A previously developed calibration has been adopted without any additional corrections being required either for the cylindrical geometry or for the different steel chemistry. This further demonstrates the versatility of laser-ultrasonics as a continuous and non-destructive bulk measurement technique that can be employed at high temperature.

The present study suggests that attenuation measurements can be used for the measurement of i) grain size evolution and ii) the start (5%) and finish (95%) times of recrystallization, thus providing a new experimental tool to observe recrystallization and grain growth following plastic deformation. These attenuation measurements provide additional and different information to laser-ultrasonic measurement of recrystallization using the velocity. The sound velocity is sensitive to the texture of the material and texture changes during recrystallization can be used to record the transformation kinetics [20-22]. In the present study, relative velocity changes of the ultrasound wave during and after the deformation are measured. The variations observed were small and of the order of the measurement precision. Thus to a first approximation, it can be concluded that very little texture change is associated with recrystallization in the present case. On the other hand, attenuation measurements of recrystallization work well provided that there are

measurable changes of the grain size during recrystallization. In particular, for hot rolling of steels, it has been observed that a limit of recrystallized grain sizes can be attained after repeated grain refinement in multi-stage deformation processes. In this special case, the initial and recrystallized grain sizes are identical and the application of attenuation-based laser-ultrasonic measurements of recrystallization may not work. Further studies will be required to explore the potential of attenuation to also provide detailed information on the recrystallization kinetics. During recrystallization, a heterogeneous, i.e. partially recrystallized, microstructure is present. Previous investigations of abnormal grain growth stages with a bimodal grain size distribution suggest that the laser-ultrasonic result is dominated by the large grains in the distribution [26]. Therefore, there may exist significant challenges in developing calibration procedures that permit to determine the recrystallized fraction between the recrystallization start and finish times using attenuation data.

An advantage that is further illustrated by the present study is that laser-ultrasonics can measure the austenite grain size evolution in low carbon steels where it is very challenging to reveal the austenite microstructure using traditional metallography. In particular for recrystallized grain sizes, metallographic data are usually restricted to carbon levels of approximately 0.1wt% or higher as the required specimen geometry for deformation necessitates sufficient hardenability to succeed with metallographic techniques. Further, laser-ultrasonics may enable to measure the evolution of the austenite microstructure in interstitial free and other ultra low carbon steels that so far have remained elusive for quantitative austenite grain size measurements.

The absolute accuracy of the laser-ultrasonic grain size measurements has been determined to be approximately 20-25%. Precision, i.e. the ability to measure small changes in grain size during an experiment, is much better, approximately 5%. At first sight, the absolute accuracy may appear low but is more than compensated by the general advantages of an in-situ technique and by the high precision. Further, the accuracy limit of metallographic investigations of austenite grain size is approximately 10% when the structure can be revealed with high quality. This accuracy drops rapidly with decreasing hardenability of the steels, i.e. with decreasing carbon content.

Evaluating the measurement accuracy for the initial grain size of the present study, there are a number of factors that may have contributed to increasing the spread of the data. For the reheating condition of 1250°C, the data (see Table II) shows a variation of initial grain size immediately after deformation from 272μm to 152μm. This deviation could be related to the different operating conditions. Smaller grain sizes are observed when the recrystallization starts within a second after deformation at higher temperatures, such as at 1100°C (Figure 3a). Further, there is a general trend of decreasing grain sizes with increasing applied strain (Figure 3b). The possible reason could be the effect of deformation on the change of sample dimension (geometry effect) or the change in grain shape which may produce a second order effect on the ultrasonic measurement of grain size. Also, it must be noted that the ultrasonic measurement of grain size utilizes a calibration that has only been validated with recrystallized microstructures showing uniform (lognormal) distributions, whereas here, the measured initial grain sizes were made on deformed microstructures. Further, the possibility of grain size gradients on the measurement plane cannot be ruled out. For example, the strain gradient in the

compression sample has been estimated to be approximately 7-10% from edge to centre. However, within the scatter that the calibration currently being used provides [18], it is reasonable to assume that the grain size gradient is a second order effect. This has been confirmed by examination of recrystallized samples for higher carbon grade steel after compression testing using similar specimen geometry, where it was possible to metallographically reveal the austenite grain structure after quenching [36]. The structures showed no evidence of a non-uniform grain size distribution. Even so, there may be potential deviations that may be acceptable if one is aware of their possible existence and takes them into account in the analysis. For example, in this paper, laser-ultrasonics could unambiguously determine the transition from recovery, recrystallization, and grain growth regimes. In addition, metallographic studies on quenched samples are time-consuming and only a few measurements are usually made to estimate a material's behavior.

Another important observation of the present research is that laser-ultrasonics provides quantitative insight into in-situ austenite grain growth after recrystallization for conditions that reflect those of industrial hot rolling, i.e. where reheating (soaking) leads to significant dissolution of precipitates. Comparing these soaking conditions with those for lower reheat temperature where no or partial dissolution takes place, as frequently associated with conventional grain growth tests, permits to critically assess previously adopted descriptions.

In general, the recrystallized grain size is frequently described by a cubic relationship with the initial grain size that is present before the deformation step such that a grain size exponent (p) of 1/3 is assumed in Eq.9 [12, 37]. This relationship can be rationalized by

assuming that new recrystallized grains form exclusively at grain boundaries and that the nucleation density on grain surface area is independent of grain size. In contrast, the present investigation reveals an apparent p value of 0.84. However, in this context, it is important to realize that the initial austenite grain size has been changed by applying different soaking temperatures and this also affects the density of precipitates. Then, the present finding suggests that different nuclei densities are present for different size distributions of precipitates. For significant dissolution, i.e. soaking at 1250°C, the nuclei density per grain boundary area is significantly lower than when precipitates are present. In the absence of precipitates, once a nucleus is formed it can readily grow along the grain boundary thereby consuming other potential nucleation sites. However, when precipitates are present, they inhibit growth along grain boundaries such that additional nuclei can form.

Previously, the softening behaviour of the present steel was analyzed neglecting the effect of precipitates, i.e. using the assumption that nuclei density is independent of the precipitate state [32]. However, the adjustable parameter to fit experimental data was the product of nuclei density and grain boundary mobility. This fit parameter appeared to be independent of the degree of particle dissolution [32]. In light of the present study the material behaviour may be interpreted as follows. For complete dissolution a low nuclei density is combined with a large effective mobility whereas for incomplete dissolution a high nuclei density pairs with a rather low effective mobility due to pinning.

This simplified approach to effectively account for pinning effects cannot be extended to describe grain growth following completion of recrystallization. Although the overall magnitude of grain growth could be replicated by introducing an effective

mobility, it would also lead to parabolic grain growth behaviour, in contrast to the experimental observations. Thus, a pinning parameter must be considered explicitly to describe grain growth. In addition, the pinning effects are much more pronounced for grain growth than for recrystallization since the driving pressures for grain growth are about three orders of magnitude lower than for recrystallization [32].

The pinning parameter to describe grain growth depends on reheat temperature and deformation condition. Although the justification of the absolute values of P is beyond the scope of this study, nevertheless their trends show a sensible pattern. As presented in Table III, the pinning parameter falls into two regions depending on the reheating conditions, i.e. P is in the order of 10^{-3}J/m^3 for reheating at 1250°C and in the order of 10^{-2}J/m^3 for lower soaking temperatures. The dissolution of previously present precipitates is either negligible or partial at lower reheating temperatures (1000-1100°C), whereas almost complete dissolution of precipitates is expected for reheating at 1250°C. Thus, there is significantly more particle pinning present for the lower reheat conditions. From an industrial perspective, the studies for the higher reheat temperature are of particular interest, as these conditions are closer to industrial practices. Also, for this reheat condition; P varies systematically as a function of the process conditions. In particular, a marked increase of the pinning parameter is found when the applied strain is increased from 0.2 to 0.5. The increase in P with strain may be an indicator of strain-induced precipitation [10-12]. In addition to the pinning parameter, the grain boundary mobility was employed as a fitting parameter to describe grain growth. This implicitly reflects that both particle pinning and solute drag are affecting grain growth. To deconvolute both effects, information on the actual particle size distribution is required and could not be

obtained with the current experimental approach. Extensive electron microscopy studies would be needed to quantify particle size distributions.

VI. CONCLUSION

Based on the current research the following conclusion can be made:

a) In-situ, non-destructive, laser-ultrasonic grain size measurement methods previously developed have been described more formally and the implicit assumptions governing the validity of those methods were explained. These measurements were extended to two new cases that of cylindrical samples and of such cylindrical samples having received hot deformations.

b) The new technique was applied successfully to the in-situ study of grain size kinetics following plastic deformation at austenitic temperatures.

c) The grain sizes measured by ultrasonics are accurate to within 20-25%. This accuracy is comparable to that of standard metallographic procedures when available. Moreover, grain size variations as small as about 5% arising from thermal processing can be detected with laser-ultrasonics.

d) Grain size evolution data measured by ultrasonics can be employed to determine recrystallization start and finish times in-situ.

e) The in-situ recrystallized grain size measurements depend weakly on deformation temperature below grain coarsening temperatures; the recrystallized grain size increases significantly with deformation temperatures when the deformation temperature is high enough so that substantial grain growth overlaps with recrystallization.

VII. REFERENCES

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Table I. Chemical composition (in wt%) of the investigated steel.

C	Mn	Si	S	P	Nb	Mo	Al	N
0.05	1.88	0.04	0.007	0.005	0.048	0.49	0.05	0.004

Table II. Experimental matrix for the laser-ultrasonic tests and the corresponding initial grain sizes for various reheat conditions

Soaking temperature [°C]	Deformation temperature [°C]	Deformation strain	Strain rate [s⁻¹]	Initial grain size [μm]
1250	1000	0.2	1.0	272
		0.3	0.1	207
			1.0	208
		0.5	1.0	172
	1050	0.3	1.0	210
	1100			152
	1150			NA*
1100	1000	0.3	1.0	52
1050				17
1000				14

NA*- Not available

Table III. Values for fit pinning parameter (P) for the grain growth model

Initial grain size [μm]	Deformation temperature [°C]	Deformation strain	Strain rate [s^{-1}]	Pinning parameter [J/m^3]
205	1000	0.2	1.0	1.5x10^{-3}
		0.3	0.1	2.1x10^{-3}
			1.0	2.0x10^{-3}
		0.5	1.0	4.5x10^{-3}
	1050			2.7x10^{-3}
	1100	0.3	1.0	1.8x10^{-3}
	1150			1.3x10^{-3}
52				1.1x10^{-2}
17	1000	0.3	1.0	1.9x10^{-2}
14				1.8x10^{-2}

Cylindrical sample
(After deformation)

Generating laser

Piston side

Load cell side

Thermocouple

Sound-wave travelling
through the sample
(generated by
generating laser)

Detecting laser

Initial grain size: 205μm
Strain: 0.3
Strain rate: 1.0/s

Figure axis labels:
- Y-axis: Grain size (μm) — values: 450, 350, 250, 150, 50
- X-axis: Time (s) — values: 0.1, 1, 10, 100, 1000

Legend text:
- Initial grain size: 205μm
- Deformation@1000°C
- Strain: 0.3
- ○ 0.1/s
- □ 1.0/s

Annotations: 5%, 5%, 1.0/s, 95%, 95%, 0.1/s

Deformation@1000°C
Strain: 0.3
Strain rate: 1.0/s