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Laser-ultrasonic austenite grain size measurements in low-carbon steels

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Abstract. Austenite grain size is an important microstructure parameter when processing steels as it provides the initial condition for the austenite decomposition that determines the final microstructure and thus properties of the steel. In low-carbon steels it is frequently difficult if not impossible to quantify the austenite grain size using conventional metallographic techniques. Laser-ultrasonics provides an attractive alternative to quantify the grain size in-situ during thermo-mechanical processing of a steel sample. The attenuation of the laser generated ultrasound wave is a function of the grain size. The present paper gives an overview on the state-of-the-art of this novel measurement technique. Using isothermal and non-isothermal grain growth tests in low-carbon steels the advantages and limitations of laser-ultrasonic measurements will be demonstrated. Further, their application for deformed samples will be presented to quantify austenite grain sizes during and after recrystallization. The in-situ measurements provide significantly new insights into the austenite microstructure evolution during thermo-mechanical processing of low-carbon steels. The implications on expediting the development of improved process models will be discussed.

Introduction

In order to develop microstructure evolution models for metals and alloys, such as grain growth and recrystallization models, it is in general required to determine material specific parameters experimentally. Another important aspect in developing these models is their validation with laboratory simulations of typical processing paths, e.g. for hot rolling or annealing. Traditionally, experimental work combines thermo-mechanical treatment of samples with post-mortem microstructure characterization using conventional metallographic techniques. This is a very labour-intensive procedure and only very limited sets of samples can be characterized. Further, there are cases, e.g. revealing austenite microstructures in low-carbon steels, where it is very challenging if not impossible to execute the metallographic characterization of the processed samples. Thus, alternative techniques must be considered in these situations. An emerging technique is laser-ultrasonics for metallurgy [1-5]. Laser-ultrasonics is a remote, continuous, and nondestructive technique that can be operated online at high temperatures for bulk observation [2, 3, 6, 7]. With this novel method it is possible to measure recrystallization and grain growth in-situ when thermo-mechanically processing the samples. In addition to obtaining microstructure data, laser-ultrasonics allows one to collect hundreds of data points rather than the few that can be obtained using conventional metallography. So far, laser-ultrasonic microstructure characterization has been conducted in dedicated laboratories where the technique has been developed using various setups of lasers. Meanwhile, laser-ultrasonics for metallurgy has reached a stage of readiness to be integrated into a user-friendly device that can be attached to a thermo-mechanical simulator thereby facilitating regular laser-ultrasonic measurements in metallurgical laboratories in the near future.

The present paper summarizes the current status of this new technique using examples of measuring austenite grain sizes during processing of low-carbon steels.

Experimental

Laser-ultrasonics. Laser-ultrasonics is the generation and detection of ultrasound using lasers. A first laser generates an ultrasound pulse, which travels inside the metal and interacts with the microstructure. Another laser coupled to an interferometer detects the ultrasound pulse at some later time. A schematic of the laser-ultrasonic measurement setup is shown in Fig. 1. The laser-ultrasonic system is attached to a Gleeble 3500 thermo-mechanical simulator. An analysis of the detected ultrasonic pulse provides the microstructure information. Ultrasound velocity is a direct measurement of elastic moduli, which depend on temperature, alloying, crystal structure, texture, precipitation and internal friction. Attenuation depends on scattering by the microstructure (i.e. by the grains) and internal friction. In the laser-ultrasonic system, a green (wavelength of 532 nm), Q-switched Nd:YAG laser with a short (5 ns) energetic (150 mJ) pulse is employed to generate a wideband ultrasound pulse by ablating a thin surface layer. The laser interferometer employs a longer pulse (50 μs with a pulse energy of 70 mJ), frequency-stabilized, infrared (wavelength of 1064 nm) Nd:YAG laser specially developed for this application. The maximum acquisition rate is 50 Hz and can thus capture rapid microstructure variations (e.g. following deformation or during rapid heating or cooling).

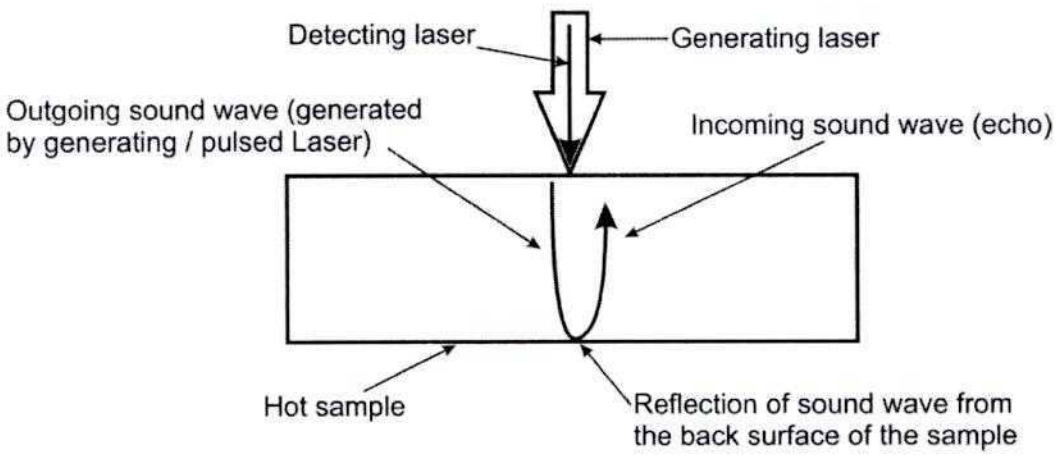

Figure 1 – Schematic of the laser-ultrasonic measurement system

Ultrasonic grain size measurements are based on the grain size dependent attenuation, i.e. [1, 8]

$$\alpha_{SC} = K(T) D^{n-1} f^{n} \tag{1}$$

where α_{SC} is the ultrasonic attenuation resulting from scattering by the grains, f is the frequency of the ultrasound wave, D is the average grain size and $K(T)$ is a parameter that depends on temperature, T. The exponent, n, falls in the range $0<n<4$ and depends on the ratio of the acoustic wave length to the grain size. Nevertheless, usually a frequency independent n-value between 1.5 and 3 can be taken [5, 9]. However, attenuation depends also on internal friction and diffraction. When an ultrasound pulse travels a distance from x_1 to x_2, the total attenuation may be written as [5]

$$\alpha = \frac{20}{x_2 - x_1} \log \frac{A(x_1)}{A(x_2)} = \alpha_{SC} + \alpha_{IF} + \alpha_{D} \tag{2}$$

where A is the amplitude of a pulse that has travelled the distance x, α_{IF} and α_D are the contributions to attenuation from internal friction and diffraction, respectively. When the sample is in the form of a thin sheet with a thickness in the order of 1-3 mm the acoustic wave is a plane wave such that $\alpha_D = 0$. Provided the grain size is sufficiently large compared to the wavelength, the attenuation is dominated by grain scattering and the contribution from internal friction can be neglected [1].

However, in general, the contribution from internal friction may not always be negligible [10]. Further, the specimen geometry may be different from the ideal sheet geometry, e.g. when deformation is involved. Then, an alternative measurement approach is required. The diffraction contribution can be determined by making a measurement at room temperature using a reference sample with the same ultrasound velocity, geometry and negligible attenuation contributions from internal friction and scattering. If possible, the reference sample is the sample that is subsequently subjected to the heat treatment or in case where the sample will be deformed the reference sample may be created by a suitable quench after completion of the thermo-mechanical treatment [5]. Further, it can be assumed that internal friction makes, to a first approximation, an unknown but frequency-independent contribution to attenuation. Then, as shown by Sarkar et al. [5], only two measurements are needed, i.e. one at the temperature of interest and the other at a reference temperature for the same propagation distance such that

$$\frac{20}{x}\log\frac{A(x,f,T_{ref})}{A(x,f,T)} = a(T) + \alpha_{sc}(f,T) = a + bf^n \qquad (3)$$

where a is a function of the contribution from internal friction. Using the frequency dependence of the measurement results, the parameters a, b and n can be obtained. According to Eq. 1, b is a function of grain size, D, and provided a suitable calibration is available the grain size can be determined. Such a calibration was developed by Kruger et al. [3] for austenite grain sizes in a wide range of C-Mn steels assuming n=3. This calibration is based on quantifying grain sizes as average equivalent area diameter (EQAD) by metallography and is used in the present paper. Further, all laser-ultrasonic (LUS) data are shown as collected to indicate the scatter of these measurements.

Materials. To illustrate the potential and limitations of this measurement technique, results from austenite grain size measurements of two microalloyed low-carbon steels will be discussed. One steel is a laboratory steel with a chemistry (in wt%: 0.05C-1.9Mn-0.05Nb-0.5Mo-0.004N) that is suitable for state-of-the-art complex-phase (CP) steels that are being developed for automotive applications. The second steel is a commercial X80 linepipe steel (0.06C-1.65Mn-0.034Nb-0.014Ti-0.14Mo-0.005N) for applications in the Arctic.

Results

Grain Growth. A conventional austenite grain growth test consists usually of heating a sample from room temperature to an austenitizing temperature where the sample is held for various times to record grain growth kinetics until a limiting grain size is attained. With laser-ultrasonics just one test provides the entire grain growth curve for a given temperature. In contrast, metallographic investigations require conducting many tests with different holding times. An example for a LUS isothermal austenite grain growth measurement series is shown in Fig. 1 for the CP steel [11]. Sheet samples with a thickness of 3 mm were heated at a rate of 5 °C/s to holding temperatures in the range of 900 to 1200 °C. As is evident from Fig. 1 the laser-ultrasonic technique nicely picks up the increase in grain size from approximately 5 μm at 900 °C to 200 μm at 1200 °C. The technique is, therefore, very suitable to record significant changes in the average grain size. A closer look at the measurement results is given in Fig. 2 for a holding temperature of 1150 °C and a comparison with

metallographic data (EQAD) is included. Comparing the two LUS measurements indicates good reproducibility. An apparent limiting grain size is reached after about 3 min of holding. Within the scatter of the laser-ultrasonic data this grain size amounts to 170±30 μm suggesting an accuracy of 20% which is comparable to traditional metallography. After 15 min holding, the measured EQAD is approximately 180±20 μm and this compares favourably with the LUS data. As shown in Fig. 4, the grain structure is rather homogeneous at this growth stage, i.e. normal grain growth takes place. However, after 2 min holding the grain size distribution is bimodal (see Fig. 4) and the "average" EQAD is much smaller than the apparent LUS grain size (see Fig. 2). Clearly, abnormal grain growth takes place at this stage, presumably due to dissolution of NbCN, and the microstructure cannot be described by an average grain size anymore. The large grains dominate the laser-ultrasonic response. In the approximation that the microstructure is a composite of a fine and a large grain component both these components will contribute to the total scattering. The attenuation caused by scattering of a material having a bimodal distribution of grain sizes can be modeled using

$$\alpha_{SC} = K(T)\left(a_1 D_1^{n-1} + a_2 D_2^{n-1}\right) f^n \tag{4}$$

where a_1 and a_2 are the volume fraction of the materials having grain sizes D_1 and D_2, respectively. Assuming that the two volume fractions are approximately equal and that n is roughly constant, the attenuation caused by scattering is dominated by the material having the larger grain size.

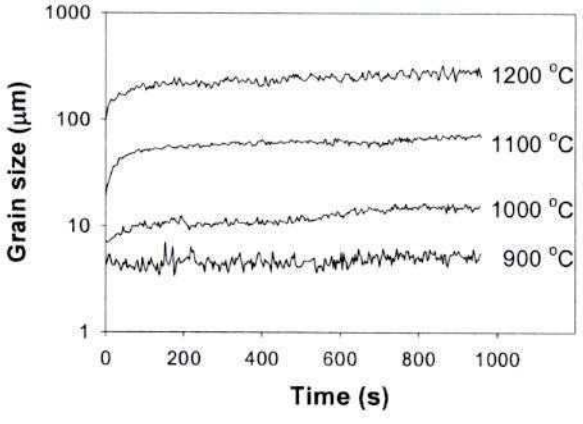

Figure 2 – Isothermal austenite grain growth in the CP steel at various temperatures

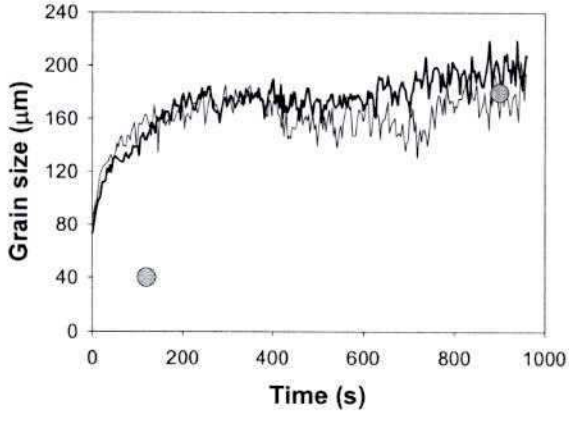

Figure 3 – Comparison of two laser-ultrasonic grain size measurements (lines) with metallographic data (symbols) for the CP steel at 1150 °C

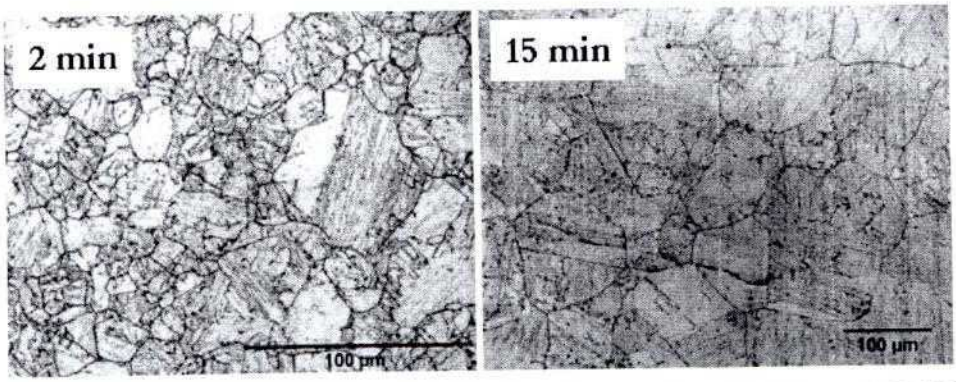

Figure 4 – Austenite microstructure of the CP steel for 2 and 15 min at 1150 °C [11]

For the X80 steel, austenite grain growth measurements were conducted with the goal to determine grain growth kinetics that are typical for the heat affected zone (HAZ) of welds. The HAZ experiences very rapid heating rates in the range of 10 – 1000 °C/s to peak temperatures that depend on the distance from the fusion zone. Previous metallographic investigations have shown that these rapid heat treatments result in normal austenite grain growth even though NbCN dissolves for sufficiently high peak temperatures [12]. For example, fine NbCN precipitates with a size of 2 nm that are present in the base metal are completely dissolved at temperatures of 1150 °C and higher for a heating rate of 10 °C/s. The laser-ultrasonic technique is particularly suitable to measure grain size evolution during rapid heat treatments as quench times that would be required for metallographic studies can be comparable to heating times and this additional time could affect the microstructure significantly. Upon reaching a peak temperature there is on average a 0.5 s effective holding time before quenching and this would be comparable to the heating time in austenite from 900 to 1400 °C at a rate of 1000 °C/s.

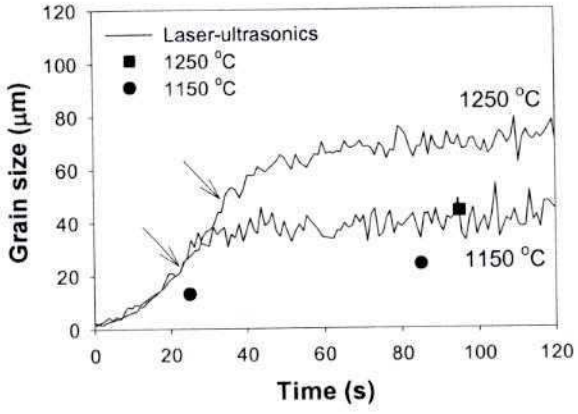

Figure 5 – Laser-ultrasonic measurement (lines) and metallographic data (symbols) of austenite grain growth in X80 steel heated at 10 °C/s to 1150 and 1250 °C. Time zero coincides with the sample reaching 900 °C, arrows indicate start of isothermal holding.

Fig. 5 shows results from laser-ultrasonic grain size measurements in the X80 steel when heated at 10 °C/s to 1150 and 1250 °C, respectively. In this graph, time is set to zero when the sample reaches 900 °C. The metallographically measured EQADs are indicated in Fig. 5 for comparison. Again the LUS data reflect the time and temperature trends of grain size evolution reasonably well and provide a wealth of data points during the heating stage with excellent reproducibility, as shown by the overlap of the two LUS curves for the initial 25 s, i.e. before reaching 1150 °C. The

quantitative agreement between metallographic and LUS grain sizes is less satisfactory even though both grain size measurements agree within a factor of 1.5. There is a tendency for the apparent LUS grain sizes to be consistently larger than the average EQAD. This observation was also made for the CP steel with finer austenite grain sizes (i.e. below approximately 50 μm). Fig. 6 provides a comparison of LUS and metallographic grain size measurements for both the X80 and the CP steel when normal grain growth takes place and the EQADs are below 50 μm. It appears that the assumed C-Mn steel calibration is not sufficiently accurate for microalloyed low-carbon steels with finer austenite microstructures. As indicated in Fig. 6, introducing a correction factor of 1.5 to the C-Mn steel calibration mitigates this problem.

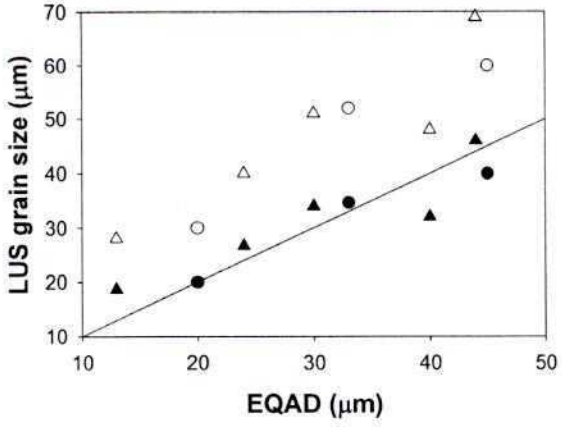

Figure 6 – Comparison of laser-ultrasonic (LUS) grain sizes with average equivalent area diameters (EQAD) obtained by metallography in CP (circles) and X80 (triangles) steels, open symbols: C-Mn-steel calibration; closed symbols: calibration corrected by factor 1.5; solid line: expected correlation.

Recrystallization and Grain Growth. In the grain growth studies sheet samples were employed such that ultrasound waves are planar waves thereby eliminating the diffraction contribution to attenuation. However, in thermo-mechanical simulations that include deformation it is impractical to use sheet samples. For example, hot deformation is often simulated by axisymmetric compression tests using a cylindrical specimen. It has been shown by Sarkar et al. [5] that cylindrical samples can be used for laser-ultrasonic measurements during and after hot deformation. For these more complex specimen geometries – after deformation the cylinder will show some barreling – it is critical to establish the simplest propagation path for the ultrasound wave. This can be accomplished by locating the excitation and detection of the acoustic pulses across the diagonal of the cylinder. Generation and detection spots have to be sufficiently small, i.e. less than 2 mm in diameter and only the first pulse travelling through the sample is analyzed. Fig. 7 shows a typical LUS measurement result using the example of a CP steel sample reheated at 1200 °C and then deformed at 1000 °C to a true strain of 0.3 at a strain rate of 1 s^{-1}. Three distinct stages can be recognized in the LUS grain size evolution curve. The initial portion replicates the grain size produced during the reheating stage (here approximately 200 μm). Using attenuation data no major microstructure changes were recorded even though recovery is expected to occur in this stage. In the second stage the LUS grain size rapidly drops (here from 200 to 100 μm), indicating that recrystallization occurs. The dashed lines in Fig. 7 describe the region between start (5% recrystallized) and finish (95% recrystallized) of recrystallization as derived from a softening model [5]. The softening model was established using independent experimental data from conventional double hit tests. There is a remarkable agreement between the predicted recrystallization start and finish times and the region of grain size refinement as recorded by laser-ultrasonics. As a result, the minimum grain size represents the recrystallized grain size – another microstructure quantity that is

challenging to determine with conventional metallography. In the third region, the grain size increases and this can be attributed to normal grain growth, i.e. one gets information on grain growth following recrystallization which is equally challenging to obtain with conventional metallographic techniques.

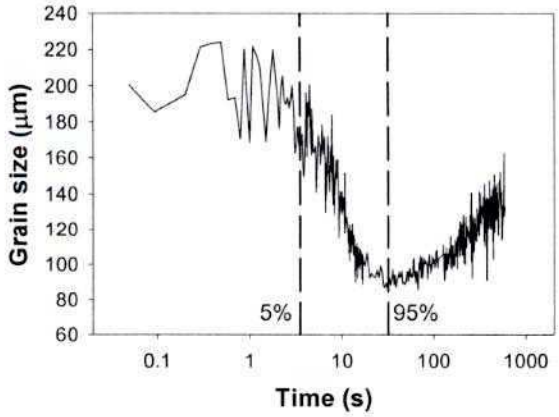

Figure 7 – Austenite grain size evolution in the CP steel during and after hot compression deformation at 1000 °C with a strain of 0.3 and a strain rate of 1 s^{-1}

Discussion

Microstructure modelling. The laser-ultrasonic technique provides continuous curves for an entire microstructure evolution process (here grain size) and many more samples and processing conditions can be analyzed than can be studied with conventional techniques. The additional experimental data will augment and expedite the development and validation of microstructure models. For example, the work by Sarkar et al. [5] on recrystallization (see Fig. 7) led to propose a revised equation for the recrystallized grain size as a function of initial grain size, applied strain and temperature. In particular, it was shown that there is a temperature dependence only for temperatures higher than 1150 °C when Nb is in solution and significant grain growth may occur during recrystallization, i.e. grains that recrystallize early can grow markedly before all grains are recrystallized. Subsequent grain growth is affected by Nb in solution and Sarkar et al. [5] showed that the grain growth rate can be described by

$$\frac{dD}{dt} = M\left(\frac{\gamma}{2D} - P\right) \tag{5}$$

where M is the grain boundary mobility, γ is the grain boundary energy and P is an effective pinning parameter. Assuming $\gamma = 0.75$ J/m^2, it was found that $M = 5.3\times10^9 \exp(-40500/T)$ (in m^4/Js) and $P = 0.002$ J/m^3 describe grain growth following recrystallization when Nb is in solution. Such information is critical for hot rolling process models but cannot be obtained from regular grain growth studies. Similar advances for grain growth models can be expected for non-isothermal conditions that are of interest for the HAZ.

Further experimental studies. In the present analysis only the attenuation data were considered and they were directly linked to the grain size. Another important information provided by the laser-ultrasonic measurement is the velocity of the ultrasound wave. Velocity information may be useful for the interpretation of grain growth data. For example, grain growth is markedly affected when precipitates dissolve. The increased amount of solutes in the matrix affects the elastic constants and this may be recorded with the velocity data. Dissolution of precipitates may lead to abnormal grain growth and this is usually accompanied by a change in texture. The ultrasound

velocity depends also on texture and carefully analyzing the velocity data may be useful to delineate abnormal grain growth stages.

Summary

Laser-ultrasonic austenite grain size measurements in low-carbon steels were reviewed. This novel technique provides unique opportunities to quantify austenite microstructures in these steels where conventional metallography is challenging. It is still required to develop more accurate grain size calibration relationships for these steels, i.e. currently laser-ultrasonic measurements have to be supplemented with a limited number of metallographic grain size measurements to benchmark the laser-ultrasonic data. Such improved calibrations will be critical to reliably use laser-ultrasonics for steels with even lower carbon content where it is not possible to reveal austenite microstructures with metallographic procedures, e.g. in interstitial free (IF) steels.
Even so, laser-ultrasonic measurements provide entire grain growth kinetic curves as long as the grain structure can reasonably be described with an average grain size. These experimental data are invaluable for developing and validating grain growth and recrystallization models. In particular, a wealth of data can be obtained along rapid heat treatment paths, for recrystallized grain sizes and grain growth following recrystallization.

Acknowledgement

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