Ultra-Fine Grained Dual-Phase Steels

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Abstract
This paper provides an overview on obtaining low-carbon ultra-fine grained dual-phase steels through rapid intercritical annealing of cold-rolled sheet as improved materials for automotive applications. A laboratory processing route was designed that involves cold-rolling of a tempered martensite structure followed by a second tempering step to produce a fine grained aggregate of ferrite and carbides as the initial microstructure for rapid intercritical annealing. The intercritical annealing step was performed with heating and cooling rates of at least 100 °C/s and a holding time of 30 s. The intercritical temperature was selected to result in 20-35% martensite in the final microstructures for C-Mn steels with carbon contents of 0.06, 0.12 and 0.17 wt%, respectively. The proposed processing routes produced an ultra-fine grained ferrite-martensite structure with grain sizes of approximately 1 μm for all three steels. The tensile strength of these ultra-fine grained dual-phase steels can be increased by up to 200 MPa as compared to coarse-grained dual-phase steels while maintaining uniform elongation values. The rather narrow processing window necessary to obtain these properties was evaluated by determining the effect of intercritical annealing conditions on microstructure evolution. Further, the experimental results were confirmed with phase field simulations of austenite formation indicating that rapid heat treatment cycles are essential to obtain fine grained intercritical austenite that leads to martensite islands with sizes of 1 μm and below in the final microstructure.

Keywords: Low-carbon steels; Intercritical annealing; Grain refinement; Mechanical properties; Phase field modelling

1. Introduction

Steel remains the most widely used engineering material with many applications particularly in the automotive and construction sectors. Enhancing mechanical properties of steels enables the reduction of cross sections in steel structures thereby leading to significant weight savings that are, for example, crucial in the design of modern vehicles. Grain refinement is a well established approach to increase strength and toughness of a material without the usage of costly alloying elements. Thus, a substantial body of work has been conducted over the past decade to develop ultra-fine grained (UFG) low-carbon steels with grain sizes of the order of 1 μm. Ferritic UFG steels have, however, limited work hardening which is a significant limitation, particularly for automotive applications. This drawback can be mitigated by combining the concept of refined microstructures with that of multi-phase structures. Currently, dual-phase (DP) steels with a conventional (i.e. comparatively coarse) ferrite-martensite microstructure are a material of choice in the automotive industry because of their favourable properties that include continuous yielding, high work hardening rate and relatively high formability. As demonstrated by a number of recent studies [1-3], refining the scale of the dual-phase microstructure increases the strength while maintaining the ductility on the levels of commercial DP steels with coarser microstructures. Unfortunately, most of these studies on fine grained DP steels employed either processing routes such as equal channel angular pressing (ECAP) that are impractical for sheet production or steels with significant alloying additions. There are just a few examples where the concept of fine grained DP steels was attempted for plain carbon chemistries [4, 5].

In particular, Azizi-Alizamini et al. [5], proposed a novel processing route to use rapid intercritical annealing to obtain a UFG DP

microstructure with improved properties in a 0.17 wt% C - 0.74 wt% Mn steel. A key element of this processing route was to develop a fine grained aggregate of ferrite and carbides before the intercritical annealing step. The goal of the present work is to evaluate the suitability of this processing route for steels with lower carbon content of approximately 0.1 wt% and below. Lowering the carbon content improves weldability and this would be advantageous for automotive applications.

2. Materials and Experimental Procedure

Two plain carbon steels (0.06 C, 0.12 C) were investigated in this study with carbon contents of 0.06 wt% and 0.12 wt%, respectively. The steels were subjected to a processing route previously developed to produce an ultra-fine grained dual-phase structure in a 0.17 wt% carbon steel (0.17 C). The three steel chemistries are given in Table 1. The 0.17 C steel is an industrially hot-rolled material whereas the lower carbon steels are laboratory materials; the 0.06 C steel was received as-hot rolled from CANMET and the 0.12 C steel as forged bars from ArcelorMittal Dofasco.

Subsequent processing in the laboratory consisted of a pre-conditioning treatment to develop a fine grained aggregate of ferrite and carbides before conducting rapid intercritical annealing cycles. Figure 1 summarizes the pre-conditioning treatment route which includes four steps. First the as-received material was austenitized and quenched to produce a martensitic microstructure. Then, the martensite structure was tempered for one

hour at 550 °C to soften the material prior to 80 pct cold rolling in a laboratory mill (roll diameter: 130 mm). Subsequently, an additional annealing was performed at 550 °C with the aim to form a fine grained ferrite-carbide aggregate. All annealing treatments were conducted in a tube furnace with Ar controlled atmosphere. The samples obtained from this pre-conditioning treatment were subjected to rapid intercritical annealing in a Gleeble 3500 thermo mechanical simulator. For this purpose test coupons of 93×12×0.7 mm were machined from the cold-rolled sheets with the longitudinal direction of the test coupon being aligned with the rolling direction. Table 2 provides the steel specific processing parameters of the employed heat treatment cycles. The tempering times after cold rolling were determined from systematic studies to optimize the formation of fine equiaxed ferrite-carbide aggregate structures in each steel. Further, it was possible to obtain an UFG DP microstructure in the 0.06 C and 0.12 C steels when reducing the heating rate into the intercritical region from 300 to 100 °C/s. This observation may be attributable to the higher Mn content of these two steels as compared to the originally studied 0.17 C steel. A detailed description of the Gleeble tests procedures is given in [5].

All microstructures were analyzed in the transverse plane of the rolled sheet, i.e. the through thickness plane perpendicular to the rolling direction.

Microstructure characterization was done by scanning electron microscopy (SEM) using a Hitachi S2300 scanning electron microscope

Table 1. Chemical composition of the investigated steels (wt%)

Steel	Fe	C	Mn	P	S	Si	Al	N
0.06C	Bal.	0.06	1.8	0.004	-	0.085	0.041	0.006
0.12C	Bal.	0.12	1.5	0.008	0.003	0.2	0.04	0.004
0.17C	Bal.	0.17	0.74	0.009	0.008	0.012	0.045	0.006

Table 2. Summary of the heat treatment cycles employed in this study

Steel	Austenitization temp./time (°C/min)	Tempering time after cold rolling (h)	Heating rate (°C/s)	Intercritical temp. (°C)	Holding time (s)	Cooling rate (°C/s)
0.06C	950/10	1	100	750	30	240
0.12C	950/10	3	100	730	30	300
0.17C	1000/30	1.25	300	750	30	1200

with a secondary electron detector. SEM micrographs were used to measure grain size and volume fraction of ferrite and martensite. Grain size measurements were based on the equivalent area diameter (EQAD) approach and at least 500 grains were analyzed. The quantitative measurements were carried out using Clemex image analysis software. Further, to quantify the resulting mechanical properties, tensile tests were conducted on samples with 12.5 mm gauge length at a nominal strain rate of 2×10^{-3} s^{-1} using a MTS servo-hydraulic machine.
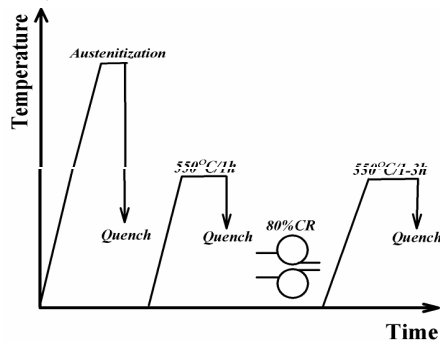
Fig. 1. Thermo mechanical process employed to develop fine grained ferrite-carbide aggregate.

3. Results

3.1. Microstructures

The pre-conditioning treatment (see figure 1) produced also in the two lower carbon steels (0.06 C, 0.12 C) a fine grained ferrite-carbide aggregate, as illustrated in figure 2. Very similar to the 0.17 C steel, the ferrite grain sizes are typically in the 1–2 μm range and carbide particles have sizes in the 30–200 nm range. The carbide size distribution is bimodal, as shown in the insets of figure 2. Larger particles with an approximate average size of ~150 nm are mainly located at ferrite grain boundaries while smaller particles with an average size of ~50 nm are distributed inside the grains. Because of the lower C contents of the 0.06 and 0.12 C steels the volume fraction of carbides is accordingly lower than in the 0.17 C steel. Further, the 0.06 C steel and to a lesser degree the 0.12 C steel display some banding in the spatial is much more homogeneous in the 0.17 C carbide distribution. The carbide distribution banding can be attributed to Mn segregation during casting and the 0.06C and 0.12 C steels have a significantly higher Mn content

(1.86 wt% and 1.5 wt%, respectively) as compared to the 0.17 C steel (0.74 wt%). Using the fine grained ferrite-carbide aggregate an ultra-fine grained dual-phase ferrite-martensite microstructure can be produced in the lower carbon steels (0.06 C, 0.12 C) similar to what had been obtained previously for the 0.17 C steel, provided a sufficiently high heating rate (here 100 °C/s) is employed to reach the intercritical annealing temperature. The intercritical temperature and holding time (see Table 2) were selected to produce a desired martensite fraction of approximately 0.2 in the 0.06C and 0.12C steels. Representative microstructures are shown in figure 3 indicating that the fine grained nature of these DP structures is similar in all three steels. In the previously studied 0.17C steel the martensite fraction is 0.35, i.e. larger than in the two lower carbon steels. As can be seen in the insets of figures 3a and 3b, the average sizes of the martensite islands are smaller in the 0.06C and 0.12C steels, i.e. 0.3 and 0.45 μm, respectively, as compared to 1.5 μm in the 0.17C steel. Further, there are remnants of banding in the distribution of the martensite islands in the 0.06C steel leading to some elongated coarser areas in the microstructure (figure 3a). The occurrence of these areas is decreased in the
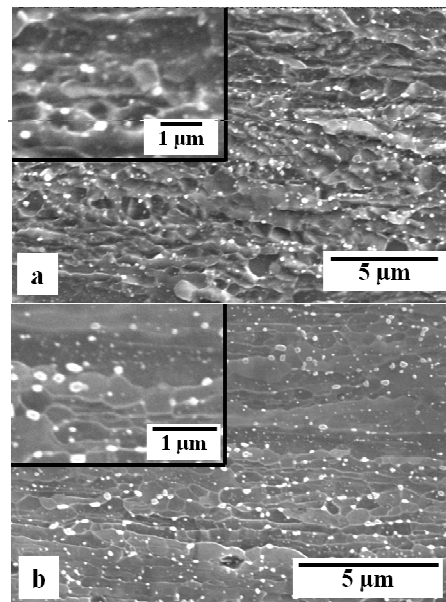

Fig. 2. Initial microstructures for intercritical annealin in (a) 0.06C and (b) 0.12C steels.

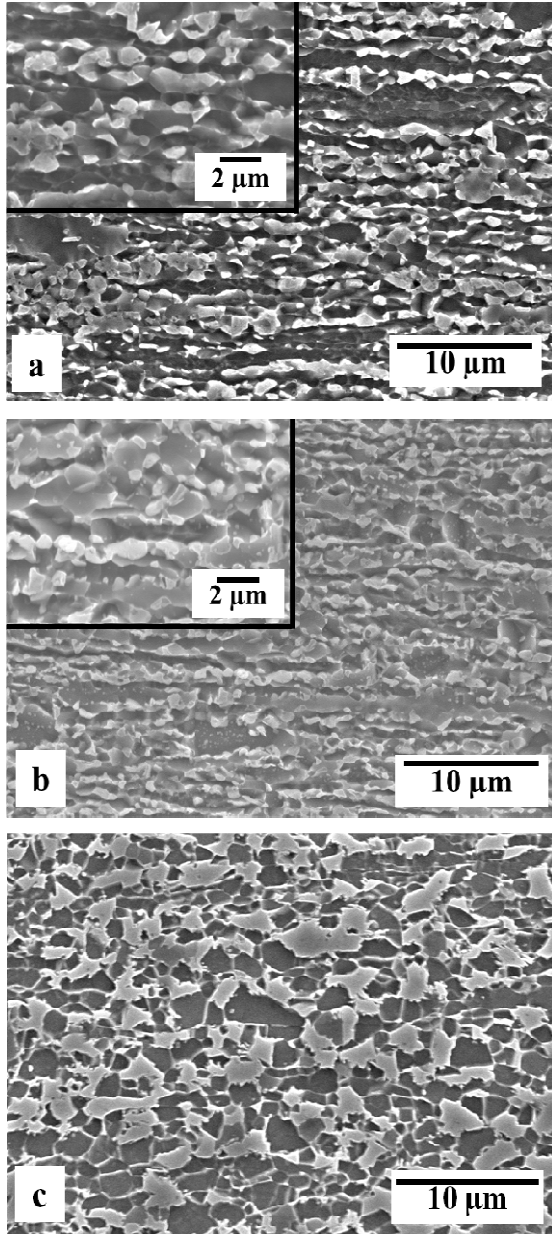0.12 C steel (figure 3b) and they are essentially not observed in the 0.17 C steel (figure 3c).

Fig. 3. Comparison of ultra-fine grained dual phase structures developed during rapid intercritical annealing in (a) 0.06C, (b) 0.12C and (c) 0.17C steels, respectively.

3.2. Mechanical properties

Figure 4 shows representative stress-strain curves for the three steels with ultra-fine dual-phase structures. In all three cases, typical DP stress-strain curves were obtained with continuous yielding and significant work hardening. The ultimate tensile strength (UTS) increases with increasing carbon content. The trend for elongation is more complex. Uniform and total elongation have their highest values for the intermediate carbon level of 0.12 wt%.

Figure 5 compares the balance of UTS and uniform elongation of the investigated UFG DP steels with data for other DP steels from the literature, as summarized in Ref. 5. For the 0.17C and the 0.12C steels this balance is better than for conventional DP steels, i.e. for a given uniform elongation the tensile strength is at least 100 MPa higher and in some cases this strength increase can reach 200 MPa. Such a property improvement is, however, not recorded for the 0.06C steel with a UFG DP structure. For this steel the strength-elongation balance remains in the range that is typically observed in conventional DP steels.

3.3. Phase Field Modelling

To further evaluate the proposed intercritical annealing routes austenite formation was simulated with MICRESS (microstructure evolution simulation software) [6] that is based on the multi-phase field approach of Steinbach et al. [7] Here, N microstructure constituents are considered and each constituent i is prescribed by its own phase field parameter ϕ_i [$i=1,\ldots,N$]. Inside constituent i, ϕ_i is equal to 1 while it is 0 outside and at the interface between two constituents, i and j, there is a gradual change of the two corresponding phase field parameters from 0 to 1 with $\phi_i + \phi_j = 1$.

The time evolution of the N phase field parameters is given by a set of coupled differential equations, i.e. [7].

$$\frac{d\phi_i}{dt} = \sum_{i \neq j} \mu_{ij} \left[\sigma_{ij} \left\{ \phi_j \nabla^2 \phi_i - \phi_i \nabla^2 \phi_j + \frac{\pi^2}{2\eta_{ij}^2} (\phi_i - \phi_j) \right\} + \frac{\pi}{\eta_{ij}} \sqrt{\phi_i \phi_j} \Delta G_{ij} \right] \quad (1)$$

where μ_{ij} is the interface mobility, σ_{ij} is the interfacial energy, η_{ij} is the interface thickness and ΔG_{ij} is the driving pressure. The phase field equations are coupled with the diffusion equations for carbon. The driving pressure for austenite formation is obtained from the linearized phase diagram as described elsewhere [8]. The interfacial

properties depend on the phase of each constituent, i.e. ferrite, austenite and cementite, respectively.

The simulations were carried out for the binary Fe-0.17wt%C alloy using the experimental rapid intercritical annealing cycle employed for the 0.17C steel. The tempering temperature of 550 °C of the pre-conditioning treatment was selected as the initial temperature. A sufficiently large mobility (i.e. 10^{-11} $m^4J^{-1}s^{-1}$) was assumed for the austenite-ferrite and austenite-cementite interfaces such that austenite formation is C-diffusion controlled. All other interfacial and diffusion parameters were taken as in previous simulations [8]. Upon reaching the Ac1 temperature of 730 °C austenite nuclei were introduced at all cementite particles existing at this point in the simulation domain. Figures 6 shows a time sequence of the simulation results. Even for a heating rate of 300 °C/s carbide coarsening takes place before the austenite start temperature of 730 °C. This limits the austenite nucleation sites to carbides located at the ferrite grain boundaries. There is also some coarsening of the austenite islands predicted during holding at the intercritical temperature of 750 °C. The simulated ferrite-austenite microstructure at the end of the intercritical annealing cycle is consistent in scale and phase fractions with the experimentally observed ferrite-martensite structures. In detail, the austenite formation rate in the C-Mn steel is lower than that predicted for the binary Fe-C alloy. To mitigate this discrepancy, a lower effective interface mobility may be introduced as proposed by Savran [9]. Nevertheless, the current simulation results indicate that a rapid intercritical heat treatment cycle is critical to obtain ultra-fine grained dual-phase structures because of rapid coarsening of carbides during heating to the intercritical region.

4. Conclusions

Rapid intercritical annealing from a fine grained ferrite-carbide aggregate is a suitable method to form UFG DP microstructures in plain low carbon steel sheets. As a result, an
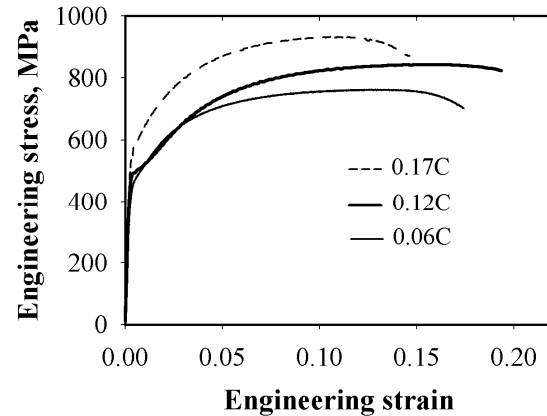

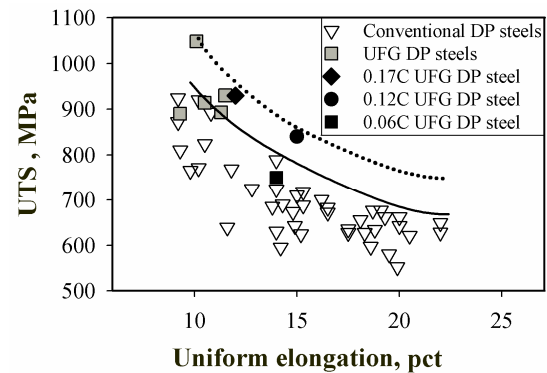**Fig. 4.** Comparison of tensile test results for 0.17 C, 0.12 C and 0.06 C steels.

Fig. 5. Balance of tensile strength and uniform elongation for different dual-phase steels (modified figure from Ref. 5 by including data for the 0.06 C and 0.12 C steels).

increased strength-elongation balance can be attained compared to conventional DP steels. In the present study, the 0.17C steel combines a tensile strength of 930 MPa with a uniform elongation of 12% whereas the 0.12C steel has a lower strength (840 MPa) but larger uniform elongation (15%). A similar property improvement has, however, not been realized for the lowest investigated carbon level of 0.06 wt% where a relatively high Mn content (1.8 wt%) leads to a banded microstructure. Thus, the present approach should be further evaluated for low carbon steels with lower manganese content (< 1 wt%).

Phase field simulations provide evidence that rapid intercritical annealing with heating rates of approximately 100 °C/s is required for the formation of UFG DP microstructures.
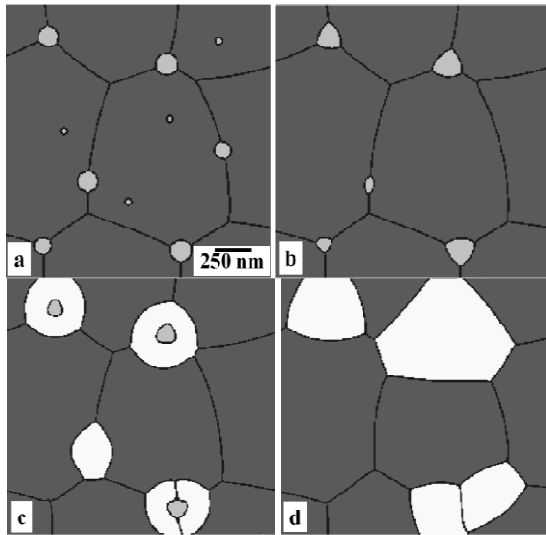
Fig. 6. Simulated microstructure evolution (dark grey: ferrite, light grey: cementite, white: austenite) in Fe-0.17C alloy during rapid intercritical annealing (300 °C/s to 750 °C and 10 s holding): (a) Initial ferrite-carbide aggregate, (b) ferrite-carbide aggregate before austenite nucleation at 730 °C, (c) early stage of austenite formation and (d) after 10 s at 750 °C.

In particular, rapid coarsening of carbides is predicted during heating reducing the austenite nuclei density thereby leading to coarser martensite islands for lower heating rates.

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