Influence of Carbon Additions on the Dynamic Recrystallization of High Purity α-Iron

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The influence of carbon content on the dynamic recrystallization mode of α-Iron containing various amounts of carbon, ranging from 5 to 200 ppm, was investigated. Uniaxial compression tests were carried out, followed by metallographic analyses by optical microscopy. All the alloys exhibit discontinuous dynamic recrystallization. Furthermore, the values of the apparent activation energies of deformation, as well as the steady state average grain sizes show that they are very sensitive to carbon additions in the range of high purities, i.e., 0–50 ppm. An interpretation of this effect is proposed on the basis of grain boundary solute segregation.

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1. Introduction

Continuous dynamic recrystallization (CDRX) is well known to occur in low carbon steels during hot deformation. However, Glover and Sellars and Tsuji et al. have shown in previous works that a transition may occur from continuous to discontinuous dynamic recrystallization (DDRX) when the material purity is increased. Indeed, the occurrence of discontinuous or continuous dynamic recrystallization, which was discussed in by Gourdet and Montheillet, depends on an equilibrium between subgrain misorientation rate and grain boundary mobility, which is strongly affected by the material purity. Such a transition has also been observed on aluminium by Tanaka et al., when decreasing the silicon and iron contents of the metal under 10 ppm.

On the basis of the preparation of high purity iron doped with controlled amounts of addition element, this work aims at investigating the influence of carbon content (5–200 ppm) on the dynamic recrystallization mode of α-Iron. A new approach of the transition observed by Glover and Sellars and, furthermore, a complete rheological investigation of these materials was carried out using hot compression tests. The deformation conditions were specified to range within the α domain of iron.

2. Experimental Procedure

2.1 Preparation of high purity iron grades with controlled carbon contents (5–200 ppm)

Commercial electrolytic iron was purified by induction melting: carbon and oxygen were eliminated by reaction with gaseous atmosphere (argon and hydrogen). Melting was performed in boat shaped crucibles, made out of silver tubes cooled by internal water flow. The solubility of Ag in Fe being virtually zero whatever the temperature, no pollution was observed. This remelting technique is described by Le Coze et al. After iron purification, the required carbon amount was introduced in the ingot by induction melting. Final carbon contents are reported in Table 1. Although the Fe–200 graded iron is beyond the limit of the α domain, the very low percentage of non α phase (also reported in Table 1) can be considered as negligible in the present study.

The ingots were hot forged to 20 mm diameter in the range and cold swagged to 9 mm diameter bars. Samples of 8 mm in diameter and 12 mm in height were then machined for hot compression testing.

2.2 Hot compression device

The servo-hydraulic machine enables hot compression from 273 to 1473 K with controlled strain rate from 10^{-3} to 10 s^{-1}. An infrared radiation furnace was mounted on this device, ensuring an homogeneity of 2 Kelvins along the sample height. Argon was used to protect samples and tools from oxidation and prevent them from pollution. Particular attention was paid to the lubrication of the specimens, because the tribological properties of high purity iron are very bad and may lead to sticking phenomena, and very large friction corrections. The lubrication procedure was as follows: polishing before each test and deposition of a boron nitride layer (spray) on the sample and the tools. The heating stage was operated with 3 thermal ramps: 105 K/min up to T_1 = (T_{testing} - 100 K), 50 K/min up to T_2 = (T_{testing} - 50 K), 10 K/min up to T_{testing}, a 10 min holding time was then applied. This thermal scheme prevents overshooting phenomena especially when reaching the testing temperature. However static recrystallization is enabled during this heating stage and strongly depends on the testing temperature. Thus a growth of the initial grain size (just before straining) may be expected when the testing temperature increases. Such variation of the initial grain size was observed and will be discussed below.

A servo-control system provides constant compression
strain rates by means of an exponential displacement signal tailored according to the initial height, the specified strain and the strain rate. Such a displacement control is required for the interpretation of the stress-strain curves, especially for deriving the rheological parameters such as the apparent activation energy, strain rate sensitivity and Zener-Hollomon parameter. After straining, the sample was quenched within about 1 second, which prevents from microstructure evolutions, in particular from static recrystallization. In the present investigation, microstructure observations are made easy by the fact that no thermodynamic transformation occurs in the quenching range (i.e. from 1073 to 293 K).

2.3 Stress-strain curves

Stress-strain curves were obtained from load and displacement measurements recorded during the tests. The ratio of the compression load to the instantaneous section first provides the apparent stress. Tribological effects are then discarded by the compression load to the instantaneous section first provides the apparent stress. Tribological effects are then discarded by the tribological effects, the apparent stress. Tribological effects are then discarded by compression. Since the influence of friction on the flow stress is limited in the investigated strain range, a Treca coefficient $\hat{m} = 0.15$ was applied for all the stress-strain curves displayed in this paper.

2.4 Microstructural investigations

After quenching the samples were cut, coated with resin, mechanically polished, and etched with 3% Nital solution. Microstructures were observed using polarized light optical metallography.

2.5 Testing conditions

Temperatures were specified within the single phase $\alpha$ domain of iron. The conditions are displayed on the Fe–C phase diagram in Fig. 1 for each of the alloys investigated. For all temperatures, a strain rate of $10^{-2}$ s$^{-1}$ was applied to the sample up to a strain of 0.7, enabling the activation energy determination for the various carbon contents (grey dots). Moreover, various strain rates from $10^{-3}$ to $1$ s$^{-1}$ were applied at various temperatures within the $\alpha$ domain for strain rate sensitivity calculations (black dots). The strain rate was always kept constant during the tests.

3. Results

3.1 Stress-strain curves

Typical flow curves obtained from the compression tests are shown in Fig. 2, displaying equivalent von Mises stress versus equivalent von Mises strain.

3.2 Occurrence of discontinuous dynamic recrystallization

Flow curves associated with discontinuous dynamic recrystallization at low strain rates and high temperatures, i.e., low Zener-Hollomon parameter ($Z$) values, exhibit typical oscillations. The latter are the result of repeated events of nucleation and growth of new grains during deformation, which lead to grain coarsening (Sakai and Jonas (8)). By contrast, in high stacking fault energy metals like aluminium or $\alpha$-iron commercial purity, where continuous dynamic recrystallization takes place, such wavy stress-strain curves have never been observed to date, whatever the $Z$ value. Within the range of high strain rates and low temperatures (high $Z$ values), DDRX is associated with grain refinement. The resulting flow curves are characterized by a single peak and attain a steady state after moderate strains ($\varepsilon \approx 1$). Still in this case, they can be readily distinguished from the CDRX ones, which exhibit a much smoother maximum and generally reach their steady states after quite large deformations ($\varepsilon > 10$). Extensive metallographic observations would be required to confirm the occurrence of either DDRX or CDRX, but it should be noted, however, that post-dynamic recrystallization might lead to wrong conclusions, especially at high $Z$ values. The stress-strain curves shown in Fig. 1 exhibit oscillations, especially at the lower strain rates. From the observation of the amplitude, frequency and number of oscillations at a constant strain rate, no specific trend indicates that iron purification favours the occurrence of discontinuous dynamic recrystallization (DDRX). Analysis of the oscillation magnitudes was nevertheless performed very carefully, because it strongly depends on the average grain size evolution between the initial state (after heating) and the steady state of deformation. In the present investigation, a large difference of initial grain size was observed between the Fe and Fe–50 grades: at 1153 K, the initial grain sizes estimated from optical metallography were 800 and 400 $\mu$m (Fig. 3, respectively). Furthermore, the final average grain size, when steady state deformation occurs, depends, of course, on the carbon content.

The different initial grain sizes are associated with static recrystallization and grain growth during the heating stage, and the temperature stabilization delay, required to ensure thermal homogeneity. The kinetics of these mechanisms strongly depend on the carbon content, since carbon atoms decrease grain boundary mobility. Quantitative comparisons of the stress-strain curves must be carried out very carefully, taking into account the specific initial state of each grade. However, the grain size evolutions observed at 1153 K and 0.01 s$^{-1}$ for a strain of 0.7 are consistent with the oscillating shapes of the stress-strain curves associated with grain growth (Table 2). Observation of the stress-strain curves confirms the occurrence of discontinuous dynamic recrystallization in low carbon (≤ 200 ppm) high purity iron. Furthermore, a weak effect of the carbon addition on the flow stress level can be noted.

In this work, purified electrolytic iron was used as a basis for preparing the various Fe–C grades. Analyses performed on this material exhibit weak amounts of cobalt (about 5 ppm), copper (about 5 ppm), and silicon (about 10 ppm). The interactions of these components with carbon and their
Influence on the recrystallization mechanism are not discussed here. They will be investigated in a future similar work starting from ultra high purity iron.

3.3 Analysis of the rheological parameters

The strain rate sensitivity of the flow stress \( m = \left. \frac{\partial \ln \sigma}{\partial \ln \dot{\varepsilon}} \right|_T \) and the apparent activation energy of deformation \( Q = \left. \frac{R}{m} \frac{\partial \ln \sigma}{\partial (1/T)} \right|_T \).
were determined from the above flow curves for each iron grade, and are reported in Table 3. Calculations were made from stress values associated with two different strain levels:

- At $\varepsilon = 0.1$, where recrystallization (whatever its type) is not yet operating, hardening and dynamic recovery are the only mechanisms that take place. At this low strain level, results must be considered very carefully, because of the influence of the material state just before straining (grain size). As described in the second part of this section, the specimens are subject to static recrystallization and grain growth during the 10 min of thermal homogenization. It is likely that the higher the deformation temperature, the larger the initial average grain size will be. Considering that static recrystallization and grain growth kinetics strongly depend on the carbon content, variations of the initial grain size may be expected for the various grades.

- At $\varepsilon = 0.6$, the deformation steady state is supposed to be reached. This assumption may be discussed for the highest strain rate, where some evolution of the stress after this strain level can still be observed. Nevertheless, the reported stress values in this case will be considered to be close to the true steady state values, considering the fairly low slope of the stress-strain curve. Since the flow stress depends only on the deformation strain rate and temperature provided that the steady state is reached, the above considerations make the values of $m$ and $Q$ at this strain level (0.6) independent of the initial state of the material.

Results obtained for the strain rate sensitivity $m$ are consistent with the literature: they exhibit in all cases an increase of $m$ with temperature, in agreement with the classical viscous behaviour predominance at high temperatures. Furthermore, Table 3 shows that $m$ is significantly higher at large strains, which might be related to the occurrence of discontinuous DRX. The apparent activation energy of the base Fe grade may be compared with the activation energy of self-diffusion $Q = 280 \text{kJ/mol}$, in $\alpha$-Fe determined by Seeger.\textsuperscript{9}

As the carbon content increases, the activation energy raises from 207 kJ/mol for Fe up to 260 kJ/mol. The value reported

Table 2 Grain size evolution during straining of the two grades Fe and Fe–50 at 1153 K and 0.01 s$^{-1}$ for a strain of 0.7.

<table>
<thead>
<tr>
<th>Grade</th>
<th>Fe</th>
<th>Fe–50</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>800</td>
<td>400</td>
</tr>
<tr>
<td>Initial estimated</td>
<td>800</td>
<td>400</td>
</tr>
<tr>
<td>average grain size (µm)</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>Final (steady state) estimated</td>
<td>1200</td>
<td>600</td>
</tr>
<tr>
<td>average grain size (µm)</td>
<td>600</td>
<td>600</td>
</tr>
</tbody>
</table>

Table 3 Rheological parameters $m$ and $Q$ of the various Fe–C grades, determined at $\varepsilon = 0.1$ and $\varepsilon = 0.6$ and various temperatures.

<table>
<thead>
<tr>
<th>Grade</th>
<th>Fe</th>
<th>Fe–50</th>
<th>Fe–100</th>
<th>Fe–200</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.1</td>
<td>0.6</td>
<td>0.1</td>
<td>0.6</td>
</tr>
<tr>
<td>$m$</td>
<td>0.1</td>
<td>0.21</td>
<td>0.13</td>
<td>0.18</td>
</tr>
<tr>
<td>973 K</td>
<td>0.14</td>
<td>0.185</td>
<td>0.14</td>
<td>0.19</td>
</tr>
<tr>
<td>1073 K</td>
<td>0.16</td>
<td>0.22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1123 K</td>
<td>0.17</td>
<td>0.22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1153 K</td>
<td>0.16</td>
<td>0.23</td>
<td>0.17</td>
<td>0.22</td>
</tr>
<tr>
<td>$Q$ (kJ/mol) at 0.01 s$^{-1}$</td>
<td>384</td>
<td>207</td>
<td>298</td>
<td>260</td>
</tr>
<tr>
<td>50</td>
<td>273</td>
<td>258</td>
<td>297</td>
<td>264</td>
</tr>
</tbody>
</table>

Fig. 4 Influence of the carbon content on the stress measured at $\varepsilon = 0.1$ ($\sigma_{0.1}$) and steady state flow stress ($\sigma_{0.6}$) at 973 K and 0.01 s$^{-1}$.

by Glover and Sellars\textsuperscript{2} was 280 kJ/mol for an alloy containing 25 ppm carbon (average value). These comparisons illustrate the relevance of the present tests, which exhibit a large effect of carbon, occurring within the range of the first few ppm additions. It is worth to note that no experimental values of activation energy had been given till now for iron with a carbon content less than 5 ppm.

Although carbon is not the only element affected by iron purification, the following analysis tries to explain its role in the mechanisms involved in discontinuous dynamic recrystallization. An interpretation of the activation energy values obtained at $\varepsilon = 0.1$ and $\varepsilon = 0.6$ can be proposed:

- At $\varepsilon = 0.1$, the microstructure of the sample strongly depends on the initial state. As described above, the initial microstructure results from static recrystallization, grain growth occurring during heating and thermal homogenization. The high activation energy for purified Fe without carbon addition may, in fact, be due to a large variation of the grain size before straining. Indeed, in a high purity metal, the grain size of a statically recrystallized microstructure increases strongly with temperature. By contrast, when the carbon content increases, the influence of temperature on the recrystallized grain size is reduced by grain boundary pinning. The initial grain size is therefore less affected by the testing temperature. In this case, the increase of activation energy is an artifact due to the prior thermomechanical history of the material: the intrinsic thermal effect on the stress level is reinforced by the increase of the initial grain size with temperature.

When the steady state is established, since discontinuous dynamic recrystallization has been observed, the stress level is significantly affected by the grain boundary mobility. The intensity of grain boundary pinning, which occurs in the Fe–C grades, lowers with increasing temperature. The increase of grain boundary mobility with temperature is therefore likely to be larger than in high purity iron, which may explain the higher values of activation energy obtained in the alloys.

This interpretation does not contradict the hardening effect of carbon observed on the stress values, because the ranges of carbon content involved by these two phenomena are quite different. The flow stress increase is strong from 0 to 50 ppm carbon, but slightly increases linearly within the range 50–200 ppm carbon, as shown in Fig. 4. By contrast, the effect of carbon on grain boundary mobility operates only at the
very low carbon contents, since no variation of the activation energy is observed between the Fe–50, Fe–100, and Fe–200 grades (Table 3). A simple interpretation of this effect can be proposed by assuming that the saturation level of a grain boundary is attained when carbon interstitials are present in one out of N lattice cells. A rough estimation of the number of carbon atoms per unit volume is then given by $\frac{3}{(NDA^2)}$, where $D$ is the average grain size and $A$ the lattice parameter. Since the number of iron atoms per unit volume is $\frac{2}{a^3}$, the ratio of the number of carbon atoms to the number of iron atoms is $\frac{3a}{(2ND)}$. Taking into account the atomic masses of the two species, this yields $36a/(112ND)$ for the mass concentration of carbon. Using $a = 0.3\text{ nm}$, $D = 10\mu\text{m}$, and assuming that $N$ ranges from 1 to 10, a critical carbon concentration from 10 to 1 ppm is obtained. This shows that carbon solutes are likely to saturate grain boundaries for volume concentrations far less than 50 ppm, which explains that grain boundary mobility and therefore the apparent activation energy remain unchanged at higher carbon concentrations.

Figure 5 shows the dependence of the steady state flow stress measured at $\varepsilon = 0.7$ on the Zener-Hollomon parameter $Z$. The three grades Fe–50, Fe–100, and Fe–200 obviously behave similarly, whereas the flow stress of pure iron is significantly larger for a given value of $Z$. This does not mean that the flow stress of pure Fe is intrinsically larger than for the Fe–C grades, but merely reflects the larger apparent activation energy of the latter at large strains (see Table 3): for a given set of strain rate and temperature, $Z$ is lower for pure iron than for the alloys, which shifts the associated line towards the left in the diagram of Fig. 4.

A similar trend is observed on the steady state grain size, which is quite similar for the Fe–50, Fe–100, and Fe–200 grades, but significantly smaller than for pure iron (Fig. 6).

These observations are of prime interest to highlight the role of carbon additions in grain boundary mobility, which is strongly decreased by the segregation of solute atoms, and is thus one of the main parameters controlling the recrystallization mechanisms.

4. Conclusions

The present paper globally confirms earlier results published by Glover and Sellars. However, the investigation was extended to a purity range below 5 ppm carbon. From compression stress-strain curves analysis and a first microstructural investigation by optical metallography, the following conclusions may be drawn:

1. The expected transition from discontinuous to continuous dynamic recrystallization with increasing carbon content was not observed in the investigated alloys, even in the Fe–200 grade. Since only continuous DRX occurs in commercial purity iron, the transition possibly occurs at higher carbon contents. Since many other elements are concerned by purification, it may also happen that the transition is mainly controlled by one of them, such as nitrogen, or substitutional elements.

2. Furthermore, the present results show that iron is very sensitive to carbon additions in the range of 0–50 ppm. This sensitivity is illustrated by the evolution of the apparent activation energy $Q$, determined either at low strains (with no DRX) or in the steady state, and by the change in steady state grain size.

3. At low strains, carbon addition decreases $Q$, since solute segregation in the grain boundaries limits grain growth prior to straining.
(4) By contrast at large strains under steady state flow, carbon addition increases $Q$, since the interaction of solutes and moving grain boundaries becomes weaker with increasing temperature. Therefore the pinning effect of carbon has opposite effects at low and large strains.

More generally, this work points out the importance of controlling the material purity even in the range of a few ppm, where many evolutions in the material behaviour and microstructure may occur.

REFERENCES