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# In situ characterization of microstructural instabilities: Recovery, recrystallization and abnormal growth in nanoreinforced steel powder

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Abstract—An in situ X-ray diffraction experiment was set up to study the microstructural evolution of a nanostructured oxide dispersion-strengthened ferritic steel produced by high-energy ball milling. Dislocation density and grain growth between 20 and 1100 °C were quantified by couplingmodified Williamson–Hall and Warren–Averbach methods. During the early stages of heating, recovery through the rearrangement of dislocations increases the coherent domain size from 23 to about 60 nm. Once the annealing temperature reaches 800 °C, recrystallization starts. Using a specific analysis of 2-D detector signal, it has been possible to grasp the occurrence of abnormal growth leading to bimodal grain size distribution with both ultrafine grains and coarser micronic grains. The grain growth kinetics upon heating were determined for both populations and separately quantified. Ultrafine grains exhibit a continuous moderate growth rate, leading to continuous recrystallization, whereas specific grains experience a rapid abnormal growth up to their final size after a short incubation time.

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#### 1. Introduction

Oxide dispersion-strengthened (ODS) ferritic steels are being intensely investigated for high-temperature nuclear applications due to their excellent creep properties and swelling resistance [1]. These assets are obtained thanks to a fine-grained body-centred cubic microstructure reinforced by a dense and homogeneous precipitation of yttrium nanooxides. The classical processing route of this material is based on powder metallurgy involving high-energy ball milling in order to obtain a homogeneous distribution of yttrium in the ferritic powder. This first step is followed by hot isostatic pressing [2-4] and hot extrusion [5]. In the most recent processing routes, the material is also regularly consolidated by field-assisted sintering [6,7]. Plastic deformation during mechanical alloying introduces a high dislocation density and deeply modifies the grain microstructure, which results in nanosized elongated grains [8]. Given the high deformation level and the small grain size, the driving force for microstructural instabilities is particularly high. Yttrium in solid solution precipitates rapidly from 600 °C under annealing. The Zener pinning force is thus also especially elevated due to the dense population of small yttrium precipitates. Having extremely high driving and retarding forces that oppose each other creates an important microstructural instability factor. The presence of an important concentration of particular pinning points, such as triple and quadruple grain boundary junctions, induced by the small grain size, will maximize the Zener force. Indeed, the smaller the grain size, the higher the number of particular points on which pinning is more efficient [9–12]. Therefore, the probability of triggering unstable phenomena like abnormal grain growth is very high.

When the material is extruded, abnormal grains elongate and are responsible for the large anisotropy of the material microstructure. This anisotropy induces detrimental mechanical properties [5] and particularly poor transverse creep strength [13], which is a key property in fuel cladding applications. Indeed, as internal pressure increases in the tube cladding with the accumulation of gas fission products, the major stress component is applied in the transverse direction. Therefore, the material faces a critical risk

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of failure and the control of the microstructure is a key issue to ensure that it can safely fulfil the role of first barrier against the release of radioactive elements. Despite its indisputable technological impact, microstructural instability, such as abnormal grain growth, of industrial nanostructured metallic materials remains only partially explored. The scarcity of such studies is primarily due to the difficulties in following the mechanisms leading to microstructural evolution: recovery, recrystallization and grain growth can be concomitant and are unpropitious to quantify in a time-resolved manner. Yet, for powder metallurgy, kinetic studies of the microstructural evolution starting from the as-milled powder and during subsequent annealing are essential to detect the beginning of critical phenomena like abnormal grain growth.

Because of its non-destructive character, X-ray diffraction (XRD) has proved to be an appropriate method for describing dislocation density and crystallite size [14-17], which are suitable microstructural features for monitoring phenomena such as recovery, recrystallization and grain growth. Nevertheless, the instability and rapid evolution of the ferritic microstructure requires the use of fast and precise in situ characterization methods. Kinetic studies have been successfully carried out by combining synchrotron XRD with fast 2-D detectors to obtain timely, wellresolved diffraction peaks [18,19]. No time-resolved study of recovery, recrystallization and further grain growth upon heating of a nanostructured ODS steel had been reported to date. In the present work, we focus on the evolution of the crystallite size and the dislocation density upon annealing. First, we report the methodology of the in situ XRD measurements and the adaptation of existing methods to improve data analysis. Since X-ray peak broadening is only sensitive to crystallite sizes below 1 µm, it is expected that this in situ investigation will deliver important information on the recovery and early stages of recrystallization of the ODS steel. These results are discussed in terms of the classical microstructural mechanisms of recovery and recrystallization. Using a special algorithm that captures individual spots of high intensity, a qualitative description of abnormal grain growth is also presented.

#### 2. Materials, experimental set-up and methods

#### 2.1. Material

A high-chromium ferritic steel powder was produced by ingot gas atomization by Aubert & Duval. The powder particles were then mechanically alloyed with submicronic yttria powder ( $Y_2O_3$ ) by Plansee SE using a high-energy attritor. The powder is representative of common industrial nanocrystalline powder widely used to process nanostructured materials. Milling conditions and microscopic evaluation of the as-milled powder are respectively reported in Refs. [8,20]. Using a focused ion beam to produce a suitable thin foil from powder particles, the nanostructure was investigated by transmission electron microscopy and automatic crystallographic orientation mapping. Most of the grains were highly deformed and the amount of geometrically necessary dislocations was estimated from the kernel average misorientation to be over  $10^{16}$  m<sup>-2</sup>. The mean grain size observed was close to 100 nm [8], a result also confirmed by scanning electron microscopy (SEM)–electron backscattered diffraction investigations [20]. The chemical composition of the milled powder was measured by electron probe microanalysis and is reported in Table 1. In this particular alloy, yttrium, titanium and oxygen are expected to form nanoparticles during hot processing [1,3,4].

#### 2.2. Synchrotron X-ray diffraction

#### 2.2.1. Experimental set-up: an in situ 2-D detection study

Various XRD experiments have shown the benefits of using the synchrotron source to study recrystallization kinetics [18,17]. This study, based on line profile analysis of in situ acquired high-resolution XRD peaks, was performed at the DIFFABS beamline of the synchrotron SOLEIL. The incident energy of photons was selected to be 19 keV ( $\lambda = 0.0653$  nm). This energy allows the optimization of the penetration depth, the number of available peaks and the peak width, each of these values being essential for the precision of the results. At this energy, the penetration depth of the beam into the powder was around 25 µm for the (211) planes. Eleven X-ray peaks can be detected in a  $2\theta$  range from 15 to 67°. Samples were tilted to 15° with respect to the axis parallel to the incident beam in order to minimize the geometrical spread of the beam onto the powder. Given these parameters, the four most intense diffraction peaks, (110), (200), (211) and (220), were located in a  $2\theta$  range at 18.5°, 26.3°, 32.3° and 37.5°, respectively. In situ X-ray diffractograms of these four peaks were recorded independently by a last-generation 2-D detector XPAD S140 composed of chips containing  $120 \times 80$  pixels with dimensions of  $130 \ \mu m \times 130 \ \mu m$  (Fig. 1).

The resulting 1-D diffractograms were obtained from a radial  $2\theta$  range of 7° and an azimuthal  $\psi$  range of 6°. Each peak was detected in a single detection sequence, leading to four different detection phases in order to centre the  $2\theta$ domain around each peak's central position. An acquisition time of 2 s per peak was chosen, providing good statistics (peak intensity of more than 1 million counted photons, giving a signal to noise ratio of more than 500 for the silicon reference sample; for the ODS samples, the resulting maximum intensity over background ratio was 85). Taking into account the XPAD displacement time from one peak position to the next, the acquisition time of the four peaks took 15 s, with an additional 10 s for returning to the initial position. The 2-D diffraction rings were then integrated over the whole  $\psi$  range for a classic measurement and over a specific  $\psi$  range to separate the ultrafine grain and coarse grain signals when an abnormal microstructure was obtained. This step was carefully performed by customized routines in order to eliminate the reconstruction artefacts that can arise during evaluation [21].

Table 1. Composition of the as-milled powder measured by electron probe microanalysis.

Element	Fe	Cr	W	Y	0	Ti	Si
Powder (wt.%) Powder (at.%)	Bal. Bal.	$14.6 \pm 0.1$ $15.1 \pm 0.1$	$0.99 \pm 0.02 \\ 0.30 \pm 0.02$	$0.16 \pm 0.02 \\ 0.98 \pm 0.02$	$0.15 \pm 0.01 \\ 0.51 \pm 0.01$	$0.32 \pm 0.02 \\ 0.35 \pm 0.02$	$0.19 \pm 0.01$ $0.35 \pm 0.01$



Fig. 1. Evolution of 2-D diffraction ring corresponding to the peak (211), from (a) as-milled powder to various annealed states (b–f) upon heating up to 1423 K. The converted plot  $I(2\theta)$  is also reported upon heating.

Using a furnace dedicated to powder heating, anisothermal treatments from room temperature up to five different soaking temperatures (T = 600, 800, 850, 900 and  $1100 \,^{\circ}\text{C}$ ) were performed in vacuum. The heating rate chosen was low enough  $(30 \,^{\circ}\text{C min}^{-1})$  to ensure that the acquisition time-induced temperature shift is only a few degrees. The powder was put onto a boron nitride sample holder that was heated by direct contact with a resistor. This experimental set-up was mounted on a rotating board that allowed the rotation around the axis perpendicular to the incident photon flux. A transparent Kapton window ensured the transmission of incident and scattered photons. A vacuum system allowed a secondary vacuum of around  $3.10^{-5}$  Pa to be rapidly obtained within the furnace chamber. Thus, considering a heating rate of  $0.5 \,^{\circ}\text{C s}^{-1}$  $(30 \text{ °C min}^{-1})$ , the temperature variation during a diffractogram acquisition (four peaks in 15 s) upon heating was less than 10 °C. Consequently, the whole diffractogram was approximated as being isothermal. Since diffractograms were measured every 25 s, the effective temperature can be estimated with an error of 13 °C.

# 2.2.2. Data analysis: from diffraction theory to metallurgical studies

XRD analyses have historically played a very important role in the study of heavily deformed materials. During the 1930s, it was thought that heavily deformed materials would produce amorphous diffraction patterns due to the high lattice distortion. However, the first diffraction experiments on highly deformed materials revealed that the matter was still ordered: the diffraction peaks were drastically broadened, but still well defined. Following this discovery, the Williamson-Hall (W-H) and Warren-Averbach (W-A) methods, both developed in the 1950s [22,23], allowed the separation of the crystallite size and the lattice strain, both of which contribute to the peak broadening. The methods have been improved repeatedly over the years [24-26]. More recently, a new step was achieved by taking into account the contrast factors of dislocations [14] (see Table 2), which depend on the elastic constants of the studied material and on the relative orientations between the line and the Burgers vectors of the dislocation and the diffraction vector [27]. This led to the so-called modified Williamson-Hall (mWH) and modified Warren-Averbach (mWA) methods.

 Table 2. Average calculated contrast factors for mixed (50/50) screw and edge dislocations [27].

Reticular plan	110	200	211	220
Contrast factor $\overline{C}$	0.141	0.2839	0.141	0.141

#### 2.2.3. Coupling the mWH and mWA methods

#### • The modified Williamson-Hall method

The Williamson–Hall method requires knowledge of the physical broadening due to microstructural evolution for each peak. In order to obtain this knowledge, the instrumental contribution was measured with a reference silicon powder mounted on a zero background sample holder. It was found that the instrumental broadening follows a Caglioti function that can be fitted by the parabolic function  $\beta_{inst} = f(\theta^2)$  [28]. The full width at half maximum (FWHM) of the physical profile was obtained using the following equation (see [29, pp. 94–126]):

$$\beta_{raw}^2 = \beta_{ODS} \beta_{raw} + \beta_{inst}^2 \tag{1}$$

where  $\beta_{raw}$ ,  $\beta_{ODS}$  and  $\beta_{inst}$  are the FWHM of the raw measured profile, and the physical profiles of the ODS and the instrument, respectively. Diffraction peaks were fitted by pseudo-Voigt functions that are defined by a convolution between Gaussian and Lorentzian functions. This function fits the diffraction pattern particularly well as the Lorentzian profile describes the grain size contribution to broadening whereas the Gaussian profile describes the strain contribution.

Since the initial mWH equation developed by Ungàr & Borbély [14], numerous evolutions of this equation have been introduced [15,17,30-32]. In order to compare our results with similar materials, we have chosen to describe the peak broadening by the equation [17]:

$$\beta_{ODS} \approx 0.9/D + \left(\frac{\pi M^2 b^2}{2}\right) \rho^{1/2} (K^2 \overline{C}) \pm O(K^4 \overline{C}^2)$$
(2)

where *K* is the scattering vector defined by  $K = \frac{2\pi sin(\theta)}{\lambda}$ , *D* is the coherent domain size (mentioned in the following as crystallite size), *M* is a dimensionless parameter linked to the dislocation density  $\rho$  and the outer cutoff radius  $R_e$  by  $M = R_e \sqrt{\rho}$ , *b* is the Burgers vector magnitude and  $\overline{C}$  is the contrast factor (see Table 2). M is sensitive to the dislocations arrangement within the material [14]. Its evolution can be summarized as follows:

- *M* > 1 indicates that dislocations are numerous and randomly distributed to form a screening strain field.
- $M \leq 1$  indicates that remaining dislocations are mostly entangled into low-angle grain boundaries (well-defined (sub) grain boundaries).
- The modified Warren–Averbach method

The mWH method relies only on the FWHM of the peaks, which is easy to measure on the experimental peak but is very difficult to calculate for a model system containing dislocations [25]. Based on the large number of variants of Eq. (2) published in the literature [15,17,30–32], we consider the values of the crystallite size and the dislocation density as only approximate. To obtain more accurate results, the mWA method, based on the asymptotic behaviour of the Fourier transform of the peak profile, was also applied. This also enables information to be obtained from more parameters – including, in particular, the evolution of the effective outer cut-off radius  $R_e$  upon heating. In this case, the instrumental contribution was corrected by dividing the Fourier coefficients of the total diffraction profile  $A_L^{raw}$  by the instrumental coefficients  $A_L^{inst}$ :

$$A_L^{ODS} = \frac{A_L^{raw}}{A_L^{instr}}$$

The instrumental coefficients were calculated by an interpolation of the neighbouring silicon peak coefficients weighted by their respective distance to the ODS peak according to the Cagliotti formula. According to Warren [22,33], the Fourier coefficients are products of size  $(A^{S}(L))$  and distortion  $(A^{S}(L))$  coefficients:

$$A_{ODS}(L) = A^{S}(L) \times A^{D}(L)$$
(3)

Finally, the mWA equation is [14,34]:

$$\ln(A_n(L)) = \ln(A^S(L))\frac{\pi}{2}\rho b^2 L^2 \ln\left(\frac{R_e}{L}\right)(K^2\overline{C}) + \frac{\pi^2}{4}Qb^2 L^4 \ln\left(\frac{R_1}{L}\right)\ln\left(\frac{R_2}{L}\right)(K^2\overline{C})^2$$
(4)

where *L* is the Fourier length, defined as  $L = n \times a_3$ , with  $a_3 = \lambda/[2(sin(\theta_2) - sin(\theta_1))]$ , *n* are positive integers,  $\lambda$  is the wavelength of X-rays and  $(\theta_2 - \theta_1)$  is the angular range of the measured peak. *Q* is directly related to the fluctuation of the dislocation density [25,30]. The quadratic part of this equation is important only when studying monocrystals or large-grained polycrystals. Therefore, for ultrafine-grained materials (such as the one studied), this term is ignored [14,15]. Plotting  $\ln(A(L))$  (at different constant values of *L*) as a function of  $K^2C$  allows the size and distortion Fourier coefficients to be obtained. According to the Warren–Averbach analysis, the crystallite size *D* is obtained from the size coefficient as:

$$\left. \frac{dA^{S}(L)}{dL} \right|_{L \to 0} = -\frac{1}{D} \tag{5}$$

The remaining strain parameters  $R_e$  and  $\rho$  were evaluated by fitting the plot of  $\left(\frac{\pi}{2}\right)\rho b^2 L^2 \ln \left(\frac{R_e}{L}\right)$  vs. L

(according to Eq. (4)). Note that the WA method describes the Fourier transform of the intensity distribution only at small values of L. Due to the inverse property of the Fourier transform, the same information can be obtained from the reciprocal space by evaluating the tails of the diffraction peaks as evidenced by the momentum method [35,36]. The method based on the FWHM of the peaks (Eq. (2)) is therefore distinctly different from the WA method since it characterizes peak broadening closer to the peak maximum, i.e. it is more sensitive to correlations between atom-pair positions corresponding to larger distances in real space. To summarize, it can be stated that the peak tails (and the mWA method) is more sensitive to average atomic structure at small real space distances, while the FWHM is more sensitive to the structure over large real space distances. Therefore, it is expected that, in the case of grain size distribution, the FWHM will be more sensitive to the population of larger grains while the mWA method will be more sensitive to the average ones. The different sensitivities of these peak parameters on the grain size is very useful for recrystallization studies, when a significant difference could indicate the onset of recrystallization and/or abnormal grain growth.

• Coupling the modified Warren–Averbach and modified Williamson–Hall methods (mWA–mWH)

Eq. (2) in the mWH method contains the unknown Wilkens parameter M, which impedes a straightforward evaluation. From the mWA analysis performed on large enough peaks, it is clearly visible that M evolves upon annealing. Its value decreases upon heating (from 2.6 to 1.7; see Fig. 2) and therefore it seems relevant to take into consideration its evolution, which accounts for the dislocation rearrangement. Nevertheless, it seems impossible to impose *M* to calculate  $\rho$  since, by definition,  $M(t) = R_e(t) \times \sqrt{\rho(t)}$ . It would therefore necessarily impose two different values of  $\rho$  within the same equation. What we propose here is to take into account the physics of the dislocation rearrangement through the outer cut-off radius parameter  $R_e$ , determined at each time step by the mWA method. In order to achieve this, the mWH method (see Eq. (2)) was adjusted by coupling it with the evolution of Re(t) upon annealing, as follows:



**Fig. 2.** Outer cut-off radius  $R_e(t)$  and dimensionless parameter  $M = \sqrt{\rho(t)}R_e(t)$  determined by the mWA method upon heating up to 600 °C (blue) and 1100 °C (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$$\beta_{ODS}(t) \approx 0.9/D(t) + \frac{\pi}{2}R_e^2(t)b^2\rho^{3/2}(t)K^2\overline{C} \pm O(K^4\overline{C}^2)$$
 (6)

This modification leads to the coupled mWA-mWH method, which partially takes into account the physics of the rearrangement of dislocations upon heating and leads to more accurate results than the classical mWH method.  $R_e(t)$  is no longer a fitting parameter but is evaluated with the mWA method. Based on the sensitivity of the FWHM to larger grains, this implies that we impose the same  $R_e$  in small and large grains. Fig. 2 shows that the approximation is reasonable up to temperatures of about 1000 °C, with the increase in *Re* in an average grain being from 23 to 35 nm only. The coupled mWA-mWH method was applied to in situ diffraction patterns acquired during several non-isothermal treatments from room temperature to 600 and 1100 °C. Unless otherwise mentioned, in the following we discuss the results mainly from the mWA method. However, to show recrystallization and/or abnormal grain growth initiation, and for the purpose of comparison, the qualitative results obtained with the coupled mWAmWH method are also shown. During the analysis, we assumed that dislocations are responsible for the entire lattice distortion and therefore ignored any chemical fluctuation. With the evolution of the dislocation system during recovery and recrystallization, only a small part of the dislocation present in subgrain boundaries remains behind in the sample, and this will be of a specific type. The average contrast factor will change in this case [15], but, due to the granular nature of the investigated sample and the large number of grains illuminated during the measurement (which means averaging over all equivalent hkl diffraction vectors), it is expected that the use of the average contrast factors will not introduce any significant errors. In the asmilled state, dislocations are supposed to be mostly concentrated on the  $\{110\}\langle 111\rangle$  dense slip systems. They are also assumed to have balanced distribution of edge and screw types [37], which is very likely in the as-milled powder but is much more questionable for the annealed samples [17,38].

#### 2.2.4. Growth of coarse grains: measurement method

Concomitant accurate measurement of the kinetics of both ultrafine grains and coarse grains is difficult to achieve since this requires getting access to the spatial distribution of grains at every time point during annealing. Recently, the growth of six individual recrystallized grains was quantified in rolled high-purity aluminium [18,39,40]. What is proposed here is to quantify the growth kinetics of abnormal grains detected in industrial ODS steels with a nonnegligible amount of second-phase particles. A number of selected grains detected as high-intensity spots in the diffraction patterns were studied upon heating. When abnormal growth occurs, the intensity in the powder diffractogram is no longer continuously distributed over the  $\psi$  range but becomes spotty (see Fig. 1). The intensity of the corresponding diffraction contribution of each grain was integrated at each time step of the annealing process. To do this, the 2-D diffraction ring was integrated in selected  $\psi$  and  $2\theta$  ranges so that only the intensity due to individual coarse grains is taken into account. Since the diffraction intensity is proportional to the volume of the selected grain, this allows the increase in volume tp be measured as a function of annealing time. The relationship between the intensity of a nanocrystalline material and that of a single grain can be given by [33]:

$$V_{grain} = m \times \frac{I_{int}^{gr}}{I_{int}^{iot}} \times \frac{\sin(2\theta)}{4\sin(\theta)} \times V_{probed}$$
(7)

where  $V_{grain}$  is the volume of the corresponding single grain, m is the crystallographic multiplicity (m = 8 for bcc structure), and  $I_{int}^{gr}$  and  $I_{int}^{tot}$  are the diffraction intensities of the single grain and the entire sample, respectively.  $V_{probed}$  is the volume of the sample probed by the X-ray incident beam. The equivalent sphere radius can be obtained as:

$$R_{eq}(t) = \sqrt[3]{\frac{3}{4\pi} \times V_{grain}(t)}$$
(8)

#### 3. Results of in situ measurements

### 3.1. Low-temperature annealing treatments ( $T = 600 \circ C$ ; $T = 800 \circ C$ )

The dislocation density and crystallite size upon heating up to 600 °C are reported in Fig. 3. Before heating, the initial nanostructure contains coherent domains with a diameter of about 23 nm, demonstrating the efficiency of highenergy attrition to produce nanostructured materials [41-43]. A considerable amount of plastic work is stored during milling. Indeed, the dislocation density is  $1.3 \times 10^{16} \text{ m}^{-1}$ which is consistent with what is observed ex situ by XRD or electron microscopy [8,20]. A first stage of rapid crystallite growth occurs at low temperatures (20–600 °C). as illustrated in Fig. 3. During these early stages of heating, annealing induces a decrease in dislocation density to about  $6.0 \times 10^{15} \text{ m}^{-2}$  (Fig. 3) and the crystallite size increases to 33 nm. After 20 min of isothermal annealing, the crystallite growth rate diminishes significantly, from  $4.0 \times 10^{-3} \text{ nm s}^{-1}$  to  $4.0 \times 10^{-4} \text{ nm s}^{-1}$ 

This can be interpreted as a transition from the annihilation of dislocations within the cells to a real increase in the crystallite size with moving crystallite boundaries. A slow increase in crystallite size is observed until the end of the soaking stage (5 h) at 600 °C, with no slope variation. The final crystallite size then is 40 nm. Meanwhile, a plateau was observed for the dislocation density at  $4.5 \times 10^{15} \text{ m}^{-2}$  (mWA). This microstructural evolution is characteristic of recovery. Note that both evaluation methods (the mWA and the newly suggested mWA-mWH methods) give identical results (for both D and  $\rho$ ) at the beginning of the tests, but they start to separate immediately before attaining the constant temperature of 600 °C. At the end of the heat treatment (16,000 s), the separation between them is still small, indicating that large and average-sized grains have nearly the same recovered dislocation structure. The corresponding dislocation densities deviate from their average value of  $5.5 \times 10^{15} \text{ m}^{-2}$  by about 20%, which is within the error of the evaluation method. However, the higher values of  $\rho$  obtained systematically from the coupled mWA-mWH method (seen for other heat treatments too) are physically non-realistic. This issue is related to the sensitivity of the FWHM to the average strain corresponding to larger real-space distances, as will be clarified in Section 4.



Fig. 3. Microstructural features upon heating up to  $600 \,^{\circ}\text{C}$  (873 K): dislocation density and crystallite size determined by the mWA the coupled mWA-mWH methods.

Fig. 4 shows that the crystallite size and dislocation density monotonously increase and decrease, respectively, with temperature. At 700 °C, the crystallite size obtained from the mWA-mWH method shows a significant sigmoidal increase. In parallel with this evolution, the decrease in dislocation density slows down significantly. This could correspond to a continuous transition from recovery to recrystallization. Indeed, the sharp decrease in dislocation density while the increase in crystallite size is slow can be attributed to the incubation time required for dislocations and sub-boundaries to rearrange and transform into proper grain boundaries. These two successive phenomena (a sharp dislocation decrease preceding a sigmoidal growth of the crystallite size) could therefore be a strong indication of an ongoing continuous recrystallization. Furthermore, the steady and moderate decrease in the dislocation density may be an indication that no discontinuous phenomena occur. At the later stage of soaking, the growth rate becomes constant for both the average and larger crystallites, with the latter being higher.

# 3.2. *High temperature annealing: the onset of a bimodal microstructure*

#### 3.2.1. 850 °C thermal treatment

During the annealing up to 850 °C, the dislocation density of the average grains (mWA method) decreases continuously from  $1.2 \times 10^{16}$  m<sup>-2</sup> to about  $2.0 \times 10^{15}$  m<sup>-2</sup>, while the crystallite size increased steadily from 23 to 37 nm (cf. Fig. 5). Larger grains (mWA–mWH data) have a very different behaviour. From 650 up to 820 °C (cf. Fig. 5), the dislocation density shows a plateau around the value of  $6.0 \times 10^{15}$  m<sup>-2</sup>. Meanwhile, the mWA–mWH crystallite size follows a sharp sigmoidal increase from 40 to 90 nm and continues to increase at higher rates throughout the soaking period. The length of the plateau in the dislocation density might correspond to the incubation time for a continuous transition from recovery to recrystallization. During the soaking stage, the growth rate of large crystallization. The arrow shows the first appearance of intense bright spot on the 2-D diffraction image, which may be associated with an abnormally grown grain. The decrease in the dislocation density during the isothermal annealing is faster compared to previous heat treatments (at 600 and 800 °C) and for average grains it reaches the value of about  $1.5 \times 10^{15}$  m<sup>-2</sup> after 1 h at 850 °C.

#### 3.2.2. 900 °C thermal treatment

During the 900 °C cycle, the signal evolution allows almost all microstructural transformations within the material to be followed step by step (Fig. 6). Once again, the recovery part of the cycle is similar to the one observed during the low-temperature annealing. The transition step between recovery and recrystallization between 650 and 800 °C is clearly confirmed; however, the length of the plateau is shorter this time. Soon after 800 °C, as observed in the two preceding cycles, the dislocation density of average grains drops to about  $2 \times 10^{15}$  m<sup>-2</sup> before that of the larger crystallite size drastically increases. It is worth noting that, in this case, the two phenomena are almost concomitant since the crystallites' sharp growth (mWA-mWH data) begins during the drop in dislocation density. This suggests an acceleration of the rate of microstructural evolution. The crystallite size increases drastically and monotonously (at a rate of  $0.4 \text{ nm s}^{-1}$ ) until abnormal grains render the results unexploitable. The dislocation density reaches  $9.0 \times 10^{14}$  m<sup>-2</sup> after the soaking period. Here, there is no transition from recrystallization to abnormal grain growth, the two phenomena occurring simultaneously.



**Fig. 4.** Microstructural features upon heating up to 800  $^{\circ}$ C (1073 K): dislocation density and crystallite size determined by the mWA and coupled mWA–mWH methods. During the first minutes of heating, the crystallite size increases sharply due to a thermal overshot. This growth is accompanied by a jump of the dislocation density. Following this change, both parameters remain constant until the temperature again reaches the peak value attained during the overshot (300  $^{\circ}$ C).



Fig. 5. Microstructural features upon heating up to  $850 \,^{\circ}C$  (1123 K): dislocation density and crystallite size determined by the mWA and the coupled mWA–mWH methods. The arrow indicates the first appearance of an abnormally grown grain.

#### 3.2.3. 1100 °C thermal cycle

Finally, a heat treatment was performed up to  $1100 \,^{\circ}$ C, the typical consolidation temperature for processing ODS steels. As illustrated in Fig. 1, the diffraction ring is no

longer homogeneous and becomes spotty upon heating up to 1100 °C. This clearly involves the occurrence of abnormal grain growth. Abnormal grains are identified as individual spots of high intensity. As emphasized in Fig. 7,



Fig. 6. Microstructural features upon heating up to 900  $^{\circ}$ C (1173 K): dislocation density and crystallite size determined by the mWA and the coupled mWA–mWH methods. The arrow indicates the first appearance of an abnormally grown grain.

the initial crystallite size and dislocation density are 22 nm and  $1.2 \times 10^{16} \text{ m}^{-2}$ , which are identical to those determined from the previously treated samples. A dislocation density plateau is once more observed (for large and average grains too) between 650 and 800 °C, definitively validating the separation between recrystallization and recovery. This is consistent with the fact that recovery is favoured over recrystallization in high stalking fault energy material and materials containing a high solute atoms content [44]. In contrast, annealing above 900 °C leads to a much more significant drop in dislocation density within the normally grown grains, which falls below  $8.0 \times 10^{14} \text{ m}^{-2}$  at 1000 °C. After 800 °C, crystallites grow very rapidly due to both continuous recrystallization and abnormal grain growth. It is interesting to note that the results between the two methods are widely divergent. Three abnormal grains were selected manually by choosing spots with high diffraction intensity. Two of the selected abnormal grains start to grow at 1080 and 1088 °C. The growth rate is much higher than that of the ultrafine grains (Fig. 8).

Indeed, once abnormal grains start to grow, they can reach their final equilibrium size at 1100 °C in less than 200 s. Given that the diffraction intensity was measured under reflection diffraction conditions and not under transmission, the error of their radius is larger than the grain radius itself. Two grains having the same volume can give rise to various diffraction intensities, depending upon the distance between the grain and the free surface, leading to an attenuation factor of between 0% to 100%. Thus the detected intensities are valuable for tracking the appearance of the abnormal grains and to observe whether they grow rapidly or not, but their volume cannot be quantified accurately. Abnormal growth occurs so rapidly that the three selected grains reach an abnormal size even before the soaking step at maximum temperature begins (cf. Fig. 8).

#### 4. Discussion

#### 4.1. Stage I: mechanisms operating at low temperature

Dislocations structures formed during cold-working of polycrystals are usually very complex. During heating, dislocations with opposite Burgers vectors can annihilate, while others can rearrange by climbing and forming lowangle subgrain boundaries, such as particular tilt boundaries [44]. In this study, the nanocrystalline powder was processed by high-energy mechanical alloving, producing a huge amount of dislocations. Due to the high stalking fault energy of the ferritic steels, these dislocations are likely to form cell structures with a typical size (determined as the initial crystallize size) of 20-30 nm. At low temperature, we have identified two different recovery mechanisms. Up to 600 °C, a monotonous decrease in the dislocation density and a monotonous increase in the crystallite size were observed. It has been shown that grain size does not evolve at this temperature [8]. This is characteristic of the part of the recovery mechanism where dislocations within cells rearrange and annihilate each other to form thinner cell walls. Therefore, the initial growth kinetics is due to dislocation annihilation within the milled powder. Nanosized cells are cleaned by dislocation rearrangement, which migrates into the subgrain boundaries of well-defined subgrains. The second part of the growth kinetics described in Fig. 3 can be considered as a second recovery mechanism with no evolution of the average grain size [8]. The growth of larger crystallites might be caused by subgrain mobility. Subgrains with a diameter D will evolve as:

$$D^n - D_0^n = c \times t \tag{9}$$

where t is the time at 600 °C, n and c are constants, and  $D_0^n$  is the diameter before subgrain growth occurred, i.e. 50 nm.



Fig. 7. Microstructural features upon heating up to  $1100 \,^{\circ}$ C (1373 K): dislocation density and crystallite size determined by the mWA and the coupled mWA–mWH methods. The arrow indicates the first appearance of an abnormally grown grain.



Fig. 8. Growth kinetics of the abnormal grains detected by the 2-D detector. The dashed line shows the annealing temperature.

A least squares method was applied to calculate the best-fit exponent n = 4.9 (Fig. 9). A wide range of values of *n* have been reported in the literature (n = 2 in high-purity iron [45]). The value found in our study is rather high, indicating a very slow subgrain growth. The increase is so slow that one would expect the mobility of the moving interfaces to be very limited. This suggests that recovery of the nanostructure at low temperature involves only homogeneous recovery, with no significant effect on grain growth. The influence of solute drag and eventual yttria clusters coming from mechanical alloying may be the cause of such a slow rate. One would expect recovery to homogeneously reduce the amount of stored energy within the deformed nanostructure. However, recovery at 600 °C involves a limited decrease in dislocation density, reaching  $4.5 \times 10^{15} \text{ m}^{-2}$ after 4 h of annealing. The amount of stored energy is therefore still important. As referred to in several studies



Fig. 9. Change in crystallite size with time at 600 °C fitted by Eq. (9).

[20,44], the dislocation density governs the driving pressure for recrystallization,  $P_V$ :

$$P_V = \alpha G b^2 \times \Delta \rho \tag{10}$$

where  $\alpha$  is a constant, G is the bulk shear modulus, b is the Burgers vector magnitude and  $\Delta \rho$  is the variation in dislocation density between neighbouring grains, which can be approximated by the mean dislocation density in the material. Consequently, the driving pressure for recrystallization is still very high even after long annealing treatment at low temperature. Since the overall solute content within the matrix is far from negligible (17 at.%; see Table 1), the solute atoms may act as obstacles and largely impede dislocation mobility [46]. At temperatures high enough to allow oxide precipitation, the precipitates have the same



Fig. 10. Comparison of the microstructural evolution during isothermal soaking time for different temperatures: (a) crystallite size, (b) dislocation density (mWA–mWH analysis).

impeding effect. The amount of stored energy available for microstructural evolution at higher temperature is therefore very high.

### 4.2. Comparison of the microstructural evolution during soaking time

When comparing crystallite sizes as a function of soaking time, two distinct evolutions can be observed (Fig. 10). At low annealing temperature the largest crystallite sizes almost reach a plateau (600°) or increase very slowly (800 °C). At high-temperature annealing (i.e. 850 and 900 °C) the larger crystallites increase very rapidly. The evolution of the dislocation density allows further increases in crystallite size. Indeed, while the dislocation density remains stable at 600 °C, it decreases steadily through 800, 850 and 900 °C. The relatively high values  $(1.0 \times 10^{15} \text{ m}^{-2})$  obtained by mWA analysis at these temperatures indicates the microstructural stability of ultrafine grains (UFGs) at high temperature.

## 4.3. Stage II: emergence of the bimodal microstructure at higher annealing temperature

• Growth of ultrafine grains

During high-temperature annealing, in the range of 800-1100 °C, the microstructural evolution is no longer confined to recovery. Nevertheless, a very clear transition is observed between recovery and the following mechanisms. Dislocation density in larger grains reaches a plateau, from 650 to 800 °C, while the crystallite size increases monotonously.

Around 800 °C, the ultrafine grain size sharply increases before returning to a steady growth meanwhile the dislocation density drops rapidly before returning to a steady decrease. This abrupt evolution is a clear sign that recrystallization occurs around 800 °C and is then followed by grain growth. An important point to note is the constant increase in the crystallite size. If new nuclei were formed, the crystallites would at least have stopped growing during the first steps of the recrystallization. Therefore, the ongoing phenomenon is very likely to be continuous recrystallization rather than primary recrystallization with grain formation. A further argument supporting this interpretation was put forward by Sallez et al. [8], who showed that grain growth involves no textural change within the UFGs regions. Nevertheless, being at such a small scale, the appearance of a few large nuclei could easily go unnoticed.

Heterogeneous grain growth was detected around 900 °C as the diffraction rings begin to form spots. The occurrence of abnormal grain growth strongly limits the contribution of nanosized grains to the diffraction signal. Indeed, the diffraction intensity is proportional to the volume of crystallite (Eq. (7)). Thus, the intensity of diffraction patterns due to UFGs is much lower than that due to coarse grains, as illustrated in Fig. 1. Despite their small size  $(0.2-1 \ \mu m)$ , UFGs are no longer quantitatively measurable and diffraction peak analysis methods reach their limits, values obtained above 0.5 µm are no longer reliable. Fig. 1 illustrates that there is still a non-negligible amount of ultrafine grains remaining in the annealed powder, as confirmed by SEM observations (Fig. 11). The remaining ultrafine grains are stable at high temperature and the dislocation density within the powder is higher than expected in annealed steels. The retained stored energy is most likely located in the ultrafine grain zones where abnormal growth did not occur. When reaching the typical recrystallization temperature, the microstructure recovery is limited to a dislocation density drop of less than one order of magnitude: from  $1.0 \times 10^{16}$  to  $2.0 \times 10^{15}$  m<sup>-2</sup>. Following Eq. (10), the driving pressure for recrystallization becomes approximately 5 MPa. Meanwhile, another study has reported a Zener pinning pressure due to nanoparticles in ODS steels close to 4 MPa [20]. Therefore, these two opposing forces are very important and this situation is very favourable for abnormal grain growth [9].

• Abnormal grain growth

Precipitation of the nanoparticles responsible for grain boundary pinning occurs between 600 and 900 °C [4]. Nevertheless, a rapid growth of grains with specific advantages (such as lower initial stored energy, higher initial grain size, lower boundary pinning force) occurs. Abnormal grain growth remains difficult to control as the factors to enable the growth of particular grains at the expense of others can be multiple and are not easily separated. In ODS steels, two parameters are more likely to influence this evolution: precipitation, which generates a pinning force, and stored energy heterogeneity. Unfortunately, both involve local measurements and are beyond the scope of the diffraction



Fig. 11. Field emission gun SEM images of the ODS steel powder after in situ annealing up to 1100 °C.

methods applied here. The abnormal grain structure is thermally stable even at high temperatures, close to the solidus temperature. As one can see with the annealed powder during the in situ measurement, the bimodal structure is still observed after heating at 1100  $^{\circ}$ C (Fig. 11).

From a processing point of view, one major conclusion can be emphasized. Whatever the consolidation time, once the process involves a temperature higher than the abnormality occurrence temperature, the resulting grain structure becomes drastically abnormal. This statement is consistent with the quantitative study on the same ODS ferritic steel consolidated by either spark plasma sintering (processing time at 1150 °C  $\approx$  5 min) or hot isostatic pressing (processing time at 1150 °C  $\approx$  3 h). A similarly heterogeneous grain size distribution was observed in the consolidated materials [20]. Indeed, the high density of nanoparticles dispersed in the ODS steels exerts an efficient Zener pinning on grain boundaries, endowing the bimodal microstructure with strong thermal stability. Consequently, this kind of abnormal grain structure might be difficult to avoid by accelerating the consolidation process kinetics. However, long annealing treatments performed just below the abnormal growth temperature may result in a significant reduction of the available energy for secondary recrystallization, thereby limiting its effects. Moreover, a complete description of the influence of precipitation on the onset of recrystallization and grain growth would allow greater control of the abnormal grain structure.

#### 4.4. Methods

#### • Comparison between methods

An important aspect of present investigation concerns the methodologies used to evaluate the crystallite size and dislocation density. At early stages of annealing, when the structure is heavily distorted, the mWA and mWA–mWH methods give identical values for the two parameters. However, as recovery and recrystallization proceed, the results given by the two methods start to differ. Although the larger crystallite size obtained from the mWA–mWH procedure can be considered reasonable in terms of the higher sensitivity of the FWHM to correlations over larger realspace distances, the higher dislocation density compared to the value given by the mWA method is physically nonrealistic. The dislocation density obtained according to the mWA–mWH method (which is supposed to correspond to larger grains) can be higher than the value found with the mWA procedure (describing average grains) by a factor of 4-5. Evidently, the rigorous theory is the latter, based on the asymptotic behaviour of the Fourier coefficients corresponding to small L. This method is more sensitive to the displacement field of single dislocations [47] and delivers the most accurate values for the average dislocation density and outer cut-off radius. Following the same train of thought, it can be argued that the FWHM becomes more sensitive not only to the larger crystallite size but also to the average strain corresponding to larger atom-pair distances. However, this strain will also be influenced by the arrangement of dislocations, which is not accounted for in Eq. (2). Since the outer cut-off radius  $R_e$  obtained from the mWA analysis changes only by a factor of 1.5 (from 23 to 35 nm, Fig. 2), it cannot be held responsible for the higher dislocation density provided by the coupled mWA-mWH procedure. We therefore consider that Eq. (2), which is widely used in the literature [15,17,30-32], is valid only for uniform dislocation structures with no subgrain size distribution. In case of structures with size distribution, the method based on the FWHM and having its origins in the Wilkens model of restrictedly random dislocation arrangement [25,48] does not deliver accurate results. Indeed, the Wilkens model subdivides the crystal into equal areas, each having the same number of dislocations [48]. In case of grains with sizes and different structures, this assumption is no more valid.

• Outer cut-off radius evolution

A characteristic parameter describing microstructural evolution is the outer cut-off radius of dislocation,  $R_e$ . The increase in  $R_e$  during recovery (Fig. 2) illustrates the transition from a random distribution of unscreened dislocations to reorganized walls. Indeed, the stored energy is reduced by dislocation migration within the crystallites into well-defined low-angle grain boundaries or subgrains. This favours the associated screening of the strain field of the dislocations. Once recovery is achieved,  $R_e$  is stable throughout the entire transition phase between recovery and recrystallization, since dislocation entanglement is unchanged and the effect of dislocations on lattice distortion is confined to within a constant crystallite size. Up to 1000 °C,  $R_e$  can be related to recovery and recrystallization: it increases at low temperature, then stabilizes while dislocations rearrange and subgrain growth is completed. Finally, it increases strongly when the microstructure recrystallizes. Above 1000 °C,  $R_e$  starts to increase exponentially. This means that it increases drastically, as the crystallite size increases until it reaches its highest possible value (Fig. 2).

#### 5. Conclusion

In situ XRD experiments have been performed to reveal the microstructural evolution process leading to the bimodal grain structure of ODS ferritic steels. Two evaluation methods sensitive to different grain populations were applied to in situ recorded diffraction images in order to capture the recovery and the onset of recrystallization and abnormal grain growth. The growth kinetics of specific abnormal grains was quantified. Regarding the selected grains only, no significant incubation time was detected. This suggests that above the onset temperature, around 850 °C, recrystallization occurs quasi-instantaneously. Therefore, annealing around this temperature seems to be of particular interest to tailor the abnormal grain ratio. Key steps of recovery and recrystallization have been identified and the microstructural evolution can be summarized as follows:

- High-energy attrition of the powder introduces a huge amount of plastic deformation, leading to a dislocation density of  $1.1 \times 10^{16} \text{ m}^{-2}$ .
- At low temperature (up to 600 °C), a first recovery step involving dislocation rearrangement induces a rapid increase in crystallite size from 20 to 50 nm.
- Between 600 and 800 °C, a second recovery step involves a slight growth of the crystallite size while the dislocation density remains almost stable.
- Around 800 °C, sigmoidal grain growth appears, while the dislocation density drops significantly, indicating continuous recrystallization.
- Above 850 °C, abnormal grains appear with no incubation time. A possible mechanism leading to this heterogeneous microstructure is the preferential growth of particular grains. It induces drastic growth and enhances grain boundary mobility, due to either stored energy or pinning force heterogeneities.

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#### Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.actamat.2014.11.051.

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