

# ESTIMATION OF DISLOCATION DENSITY IN BAINITIC MICROSTRUCTURES USING HIGH RESOLUTION DILATOMETRY

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## Abstract

It is possible by means of high resolution dilatometry together with a model based on isotropic dilatation and atomic volumes, to estimate the dislocation density introduced in the microstructure as a consequence of the isothermal decomposition of austenite into bainitic ferrite. The relatively high dislocation density associated with this microstructure is attributed to the fact that the shape deformation accompanying this displacive transformation is accommodated by plastic relaxation.

**Keywords:** bainitic steels; phase transformation; high resolution dilatometry ;dislocations

Dilatometry is a powerful technique widely used in steels to determine critical transformation temperatures, Curie temperatures and thermal expansion coefficients of phases as ferrite and austenite [1]. More recently with the development of high resolution dilatometers, the possibilities have expanded to the study of other phenomena such as texture changes and rearrangement and annihilation of dislocations associated with recovery and recrystallization processes [2]. Therefore it is reasonable to assume that the same technique will be also sensitive to the introduction of dislocations as a consequence of a phase transformation. To probe this point carbide free bainitic microstructures obtained by the isothermal decomposition of austenite at low temperatures have been selected.

Carbide free bainitic microstructures are regarded in this work as the product of the isothermal decomposition of austenite into a mixture of bainitic ferrite plates and carbon enriched austenite. Cementite precipitation is avoided by the judicious use of silicon as an alloying element. Nowadays there are plenty of well established experimental data that confirm that nucleation of bainite takes place under paraequilibrium conditions (only C diffuses) and its growth is martensitic in nature, meaning that unlike ferrite or pearlite, bainite grows by a deformation of the parent austenite without any diffusion of atoms

[3,4]. The deformation causes a change in the shape of the transformed region, consisting of a large shear and a volume expansion [4]. As bainitic transformation takes place in a temperature range where the shape change cannot be accommodated elastically, the plastic deformation that is driven by the shape change causes the accumulation of dislocations, increasing in number as transformation temperature decreases [5,6]. The dislocation density is important in determining the mechanical properties or tempering resistance of bainitic microstructures [4,7,8]. Techniques as transmission electron microscopy and analysis of X-ray peak broadening have been already used successfully for the determination of the dislocation density [8], in this work we explore the possibility of attaining similar measurements by means of high resolution dilatometry.

The alloy selected for this work (0.78C-1.6Si-2Mn-1Cr-0.24Mo-3.96Co-1.65Al wt.%) has been designed on the basis of phase transformation theory alone [4], in order to obtain a nanostructured carbide free bainitic and dislocated microstructure after isothermal heat treatment at low temperatures. The experimental bainite and martensite start temperatures,  $B_s$  and  $M_s$ , were determined as  $410\pm 15^\circ\text{C}$  and  $188\pm 2^\circ\text{C}$  respectively. Therefore the selected transformation temperatures were 375-350-300 and  $250^\circ\text{C}$ . Detailed information on the metallurgical design of this variant of bainitic nanostructured steel and of its heat treatment is given somewhere else [9-11].

A Bähr 805D high resolution dilatometer was used to track the isothermal decomposition of austenite into bainitic ferrite on samples of 5 mm diameter and 10 mm length. In the present experiments, each sample is heated by induction to a temperature of  $950^\circ\text{C}$  for 15 minutes and subsequently cooled down to the isothermal temperature at a rate of  $10^\circ\text{C/s}$ , the whole process was carried out in vacuum of  $5\times 10^{-6}$  mbar. Finally samples were Ar quenched to room temperature. The length change is recorded by a Linear Variable Displacement Transducer with a resolution of  $0.05\mu\text{m}$ . Giving the low transformation temperatures selected for this work, and keeping in mind the small expansion coefficient of quartz (push rods),  $0.5\times 10^{-6}^\circ\text{C}^{-1}$ , when compared with the expansion coefficient of steel, approximately  $10\times 10^{-6}^\circ\text{C}^{-1}$ , it is safe to conclude that the contribution of the push rods to the measured length is negligible.

Standard metallographic techniques were used to prepare appropriate samples for observation under a Jeol JSM-6500F field emission gun scanning electron microscope operating at 15 kV and a JEOL JEM-200CX transmission electron microscope operated at 200 kV.

Quantitative X-ray diffraction analysis was used to determine the fraction of retained austenite and bainitic ferrite,  $f_\gamma$  and  $f_\alpha$ , and its respective lattice parameters  $a_\gamma$  and  $a_\alpha$ . Experiments were performed in a SIEMENS D 5000 X-ray diffractometer using unfiltered Co  $K\alpha$  radiation, a scanning speed ( $2\theta$ ) less than 0.3 degree/min and operated at 40 kV and 30 mA. A more extensive description of sample preparation and the methods used to obtain the volume fraction of retained austenite and bainitic ferrite as well as accurate values of their respective lattice parameters could be found in [7,12].

Moreover, retained austenite composition was calculated making use, as described in [13], of the relationship between lattice parameter and chemical composition reported by Dyson and Holmes [14], this expression has been selected for being the most complete in terms of the influence of different elements in the austenite lattice parameter and its use has been validated by the authors in several works [7,9,15,16], where it has been probed the good level of agreement with atom probe tomography measurements [17].

For the sake of results comparison, X-ray data from the microstructure were also analyzed in terms of peak broadening to study the microstrains associated with the introduction of dislocations during bainitic transformation as detailed in [7,8,18].

### **Transformation**

Figure 1 shows the dilatational curves associated with the bainite transformation at the selected isothermal transformation temperatures. The maximum extent of transformation increasing as it does the undercooling below the  $B_S$  (410°C). Metallographic analysis showed that the general microstructure consists of a mixture of solely two phases, plates of bainitic ferrite and retained austenite, as described in [9,16] and illustrated in Table 1. The amount of bainitic ferrite present at the microstructure increases as the transformation temperature decreases, which is in agreement with the degree of transformation shown in Figure 1 and also with the principles of the bainitic transformation theory used for the design of this steel [4].

Figures 2 (a) and (b) shows selected micrographs revealing the fine size of the developed microstructures. The darker long and slender features are the plates of bainitic ferrite, and the lighter phase found as really thin films and more blocky type features correspond to retained austenite. The scale of the microstructure is consequence of the increased strength of the parent austenite at low temperatures and the increase in the magnitude of the free energy change accompanying transformation [19].

Extensive scanning and transmission electron microscopy examination of the samples did not reveal any cementite, as it was expected when adding Si to the chemical composition, Figure 2 (b).

## Analytical Transformation Strains for Austenite Decomposition

Dilatometry is a powerful technique for the study of phase transformations in steels, since density change resulting from phase transformation gives rise to an observable dilatation different from the thermal expansion effect. Under the assumption of isotropic dilatation behavior, the volume change ( $\Delta V = V_f - V_0$ ) with respect to the initial volume ( $V_0$ ) is related to the relative length change ( $\Delta l/l_0$ ) by

$$\frac{\Delta V}{V_0} = \left(1 + \frac{\Delta l}{l_0}\right)^3 - 1 \approx 3 \frac{\Delta l}{l_0} \quad (1)$$

Since the value of ( $\Delta l/l_0$ ) is very small, the square and cubic terms of  $\Delta l/l_0$  can be neglected. The right-hand side term in the equation above,  $3(\Delta l/l_0)$ , can be directly measured by dilatometry, while the left-hand side term,  $\Delta V/V_0$ , represents the volume change, which can be calculated based on the lattice parameters of the existing phases and is directly related to the fraction change. Therefore in principle it should be straight forward to theoretically obtain the total  $\Delta l/l_0$  consequence of the bainitic transformation at each temperature.

### Initial state ( $V_0, \gamma_0$ )

The initial state is considered as that of austenite ( $\gamma_0$ ) at the isothermal transformation temperature previous to the beginning of transformation. Thus, austenite chemical composition is equal to that of the bulk material. The atomic volume of austenite as a function of its lattice parameter is given by  $V_0 = V_{\gamma_0} = a_{\gamma_0}^3 / 4$  since the unit cell of austenite contains 4 Fe atoms. For the calculation of the room temperature austenite lattice parameter with a chemical composition equal to that of the bulk material we assume Dyson and Holmes expression [14].

Finally, the lattice parameter of untransformed austenite at the transformation temperature T is calculated from the room temperature (25°C) lattice parameter using the thermal expansion coefficient  $e_\gamma$

$$a_i(T) = a_i(25^\circ\text{C}) (1 + e_i (T - 25)) \quad (2)$$

where T is the temperature in °C, and  $a_i$  and  $e_i$  represent the lattice parameter and thermal expansion coefficient of phase i, in this case austenite ( $\gamma$ ).

### Final state ( $V_f, \gamma^+ + \alpha$ )

At the end of the isothermal heat treatment at a given T, the microstructure consists solely of carbon enriched austenite ( $\gamma^+$ ) and bainitic ferrite ( $\alpha$ ). The level of C enrichment in austenite is high enough to retain it at room temperature after cooling from the isothermal heat treatment temperature. Martensite formation was neither detected by dilatometry nor by metallographic observation, thus, the atomic volume of the sample can be determined by the fractions of the phases present multiplied by their atomic volume,  $V_f = f_\gamma V_{\gamma^+} + f_\alpha V_\alpha$  where  $V_{\gamma^+} = a_{\gamma^+}^3 / 4$  and  $V_\alpha = a_\alpha^3 / 2$ ,  $f_i$  is the fraction of phase i (as listed in Table 1).

Therefore, the lattice parameters of austenite and ferrite at the end of the heat treatment at a temperature T can be derived from those obtained at room temperature by means of X-ray, Table 1, and then applying eq. (1). According to the methodology just described the only unknown variables to obtain the theoretical ( $\Delta l/l_0$ ) associated with  $\gamma_0 \rightarrow \gamma^+ + \alpha$  transformation are the thermal expansion coefficients,  $e_i$ , of austenite and ferrite. For the case of austenite,  $e_\gamma$  was derived from the austenite region of the dilatometric curves obtained from an experiment where a sample was heated at 0.2°C/s up to 1100°C and immediately cooled down at the same cooling rate, the same experiment was conducted but with a reference sample of pure Pt, then the data from the steel experiment were corrected with the Pt one. By following this procedure we ensure that the corrected dilatometric signal corresponds exclusively to processes taking place in the steel sample. The results thus obtained were 22.6 and 23.0  $\times 10^{-6} \text{ }^\circ\text{C}^{-1}$  during heating and cooling respectively. The problem arose when trying to obtain  $e_\alpha$ , as the alloy has a high C content, during cooling austenite transformed to pearlite with some ferrite, which made impossible the determination of  $e_\alpha$ . Therefore, it was decided to determine the strain associated with the bainitic transformation using sets of  $e_\gamma$  and  $e_\alpha$  reported in the literature [20-22]. Note that in the case of ref 22 the value of  $e_\gamma$  varies with the C content of austenite following the expression  $e_\gamma = (24.9 - 0.5 C_\gamma) \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ , where  $C_\gamma$  is the atomic percent of C in austenite, therefore it is possible to provide with a value of  $e_\gamma$  for the bulk composition, and for the different C levels measured after treatment at the different isothermal temperatures, as reported in Table 1.

Figure 3 allows us to compare the  $\Delta l/l_0$  values obtained from the dilatation curves, Figure 1, and those obtained theoretically following the procedures just described for the different set of thermal expansion coefficients selected. It is obvious that regardless of the exact values of  $e_i$  used, the experimental values of the strain associated with bainitic transformation are always bigger than that obtained theoretically, and the difference increases as the transformation temperature decreases.

As in the case of martensite, growth of bainite is accompanied by a shape change of the transformed region. As bainitic transformation takes place in a temperature where the shape change cannot be accommodated elastically, the plastic deformation that is driven by the shape change causes the accumulation of dislocations, increasing in number as transformation temperature decreases [5,6]. Figure 2 (d) illustrate the great deal of dislocation debris perceptible in the microstructure, which has been identified as glissile pure-screw dislocations [4,23]. The micrograph also reveal the nano character of the developed microstructures.

According to these premises it is reasonable to assume that the difference  $(\Delta l/l_0)_{\text{exp.}} - (\Delta l/l_0)_{\text{theo.}}$  can only be identified as microstrain,  $\varepsilon$ , i.e non-uniform strains in the microstructure consequence of the dislocation density,  $\rho$ , inherent to the nature of the bainitic transformation. In general,  $\varepsilon$  is proportional to the square root of the dislocation density  $\rho$ , and according to a dislocation density model [24] microstrain can be converted to  $\rho$  by  $\rho = k \varepsilon^2 / F b^2$ , that allow us to estimate the dislocation density introduced in the microstructure, where the Burgers vector  $b = 0.25 \text{ nm}$ , and  $F = 1$  and  $k = 12 A$  ( $A = \pi/2$ ) [24,25]. The necessary thermal expansion coefficients values for the calculations were selected as those in ref. [22], because  $e_\gamma$  with the bulk composition,  $23.19 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ , shows an excellent agreement with that measured experimentally by the authors in this work,  $23.0 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ .

Finally, in Figure 4 is presented the estimation of the dislocation density at the different transformation temperatures following the procedures just described. As expected, results show a tendency to increase the dislocation density from  $3.77 \pm 2.05 \times 10^{14} \text{ m}^{-2}$  to  $4.20 \pm 1.90 \times 10^{15} \text{ m}^{-2}$  as the transformation temperature decreases from 375 to 250°C, and in all cases are well above those reported for allotriomorphic ferrite,  $3.7\text{-}5.0 \times 10^{13} \text{ m}^{-2}$ , [26,27]. The same dislocations densities have been calculated by means of X-ray peak broadening analysis, as detailed in [7,8,18], and although slightly higher, results obtained by both techniques are in reasonable good agreement. In general terms the results here presented are within those measured by other authors for bainitic and martensitic microstructures [28-30], and those obtained by us using X-ray peak broadening methods in a similar set of bainitic steels also treated isothermally [8].

It has been shown that high resolution dilatometry is sensitive to the microstrain (i.e. dislocation density) introduced in the microstructure during bainitic transformation as a consequence of its displacive nature. This works proposes a method where using the dilatometric signal and assuming isotropic dilatation and atomic volumes of the phases present, austenite and bainitic ferrite, ultimately leads to an

estimation of the dislocation density, showing good agreement with those reported by other authors and techniques.

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## Table

Table 1. Quantitative X-ray data of the microstructures at room temperature, where  $f_{\alpha}$  is the fraction of bainitic ferrite being the fraction of austenite  $f_{\gamma} = 1 - f_{\alpha}$ ,  $a_i$  are the lattice parameters and  $C_{\gamma}$  is the austenite C content.

Transformation Temperature	375°C	350°C	325°C	300°C	250°C
$f_{\alpha}$ ( $\pm 0.02$ )	0.57	0.55	0.64	0.61	0.72
$a_{\gamma}$ / nm ( $\pm 0.0001$ )	0.3621	0.3629	0.6334	0.3635	0.3632
$a_{\alpha}$ / nm ( $\pm 0.0001$ )	0.2872	0.2870	0.2871	0.2871	0.2874
$C_{\gamma}$ / at. %	4.36	5.25	5.87	6.00	5.627
$C_{\gamma}$ / wt. %	1.00	1.21	1.37	1.39	1.31

## Figures

Figure 1. Dilatational curve showing longitudinal strain associated with the bainite transformation at different transformation temperatures.

Figure 2. Scanning and transmission electron micrographs of the microstructure after transformation at (a) 300°C and (b) 250°C.

Figure 3. Comparison of the theoretical and experimental strains ( $\Delta l/l_0$ ) associated with the bainitic transformation at different temperatures.

Figure 4. Calculated dislocation density and also by means of X-ray peak broadening analysis, B stands for bainitic microstructure. Also included values reported by other authors for martensitic (M) and bainitic microstructures. ref. 29 refers to an empirical model to determine the dislocation density as a function of transformation temperature in displacive transformations, i.e. bainite and martensite.