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AN INVESTIGATION OF THE MECHANISM OF BAINITE TRANSFORMATION IN EXPERIMENTAL 0.2C-1V-2Mn STEEL

ABSTRACT

The paper presents an investigation of the mechanism of bainite transformation in 0.2C-1V-2Mn steel using high speed dilatometry and TEM backed by thermodynamic analysis. Obtained results confirm the incomplete reaction phenomenon with the cessation of the bainite transformation well before paraequilibrium is achieved. These experimental data indicate that bainitic ferrite forms by a displacive transformation mechanism, but soon afterwards, excess of carbon is partitioned into the residual austenite. The results are discussed in terms of the mechanism of bainite transformation.

Key words: *mechanism of bainite transformation, lower, upper bainite, carbon partitioning*

INTRODUCTION

The mechanism of bainite transformation and carbon concentration in the residual austenite at the termination of the bainitic reaction in steels remain still controversial and involves two general concepts, that bainitic reaction occurs via a diffusional mechanism [1], and that bainitic reaction occurs in a displacive mechanism as far as the substitutional elements are concerned [2,3,4]. In case of bainitic transformation, reaction ceases before the parent phase has completely transformed what is known as an incomplete reaction phenomenon. It means that at any temperature below B_S and in the absence of any interfering secondary reactions only a limited quantity of bainitic ferrite forms before the reaction terminates. The role of carbon in the growth of bainite, is difficult to resolve directly, because even if the ferrite is supersaturated during growth, the relatively high transformation temperatures provide an opportunity for the excess of carbon to diffuse rapidly into residual austenite before any experimental measurements can be made [5,6,7,8]. However, there is indirect method of determining the carbon concentration of bainitic ferrite during growth, e.g. dilatometry method. This paper attempts to investigate the mechanism of the bainite transformation in 0.2C-1V-2Mn steel and to clarify the bainitic ferrite formation mechanism from the thermodynamic point of view.

EXPERIMENTAL PROCEDURES

A high-speed Adamel Lhomargy LK-02 dilatometer was used to establish change of length ($\Delta L/L$) during isothermal bainitic transformation. In order to ensure rapid cooling ($\sim 300\text{Ks}^{-1}$) from austenitizing temperature (1000°C , 10 minutes), the specimens were 13mm in length and 1.1mm in diameter. Lattice parameter measurements were carried out using a X-ray diffractometer with Fe-filtered CoK_α radiation. The precision ferrite lattice parameter determination included a knowledge of the angular positions of the (110), (200), (211) and (220) peaks. The data were analysed using a Taylor-Sinclair function to extrapolate the values of the ferrite parameter to angular position of $\Theta=90^\circ$. The linear expansion coefficient of ferrite (e_α) was determined by annealing a specimen at 650°C for 30 minutes to decompose any retained austenite and then recording the change of length during slow cooling. The linear expansion coefficient of austenite (e_γ) was measured after cooling from temperature of 1100°C while the specimen was in the single γ phase field. Determination of the linear expansion coefficients was carried out in a UBD Leitz-Wetzlar dilatometer. The various morphologies of bainite were distinguished by optical and electron microscopy.

The specimens for optical microscopy were etched using 2% nital. Thin foils for transmission electron microscopy (TEM), were prepared from discs slit from heat-treated specimens used in the metallographic techniques. The discs were mechanically thinned to $50\text{ }\mu\text{m}$ and were electropolished until perforation occurred in a twin jet polishing unit containing an electrolyte of 5% perchloric acid, 25% glycerol and 70% ethyl alcohol mixture solution. The electrolyte temperature was maintained around -10°C , the polishing potential was 55 V at a current of 30 mA. Thin foils were stored in ethanol and subsequently examined in a Tesla BS-540 electron microscope at an operating voltage of 120 kV.

CONVERSION OF DILATOMETRY DATA

For the transformation of austenite into a mixture of bainitic ferrite and carbon enriched residual austenite and assuming isotropic strain the relationship between dimensional changes ΔL and volume changes ΔV is given by the equation [9]:

$$\frac{\Delta V}{V} \approx 3 \frac{\Delta L}{L} \quad (1)$$

Where L is the specimen length at the transformation temperature. The lattice parameters of bainitic ferrite and untransformed austenite at the reaction temperature, a_α and a_γ , can be calculated from the values of lattice parameters at ambient temperature (25°C , 298K) using the linear thermal expansion coefficients e_α and e_γ :

$$a_\alpha = a_{o\alpha} [1 + e_\alpha (T - 298)] \quad (2)$$

$$a_\gamma = a_{o\gamma} [1 + e_\gamma (T - 298)] \quad (3)$$

where T is the absolute temperature. The room temperature lattice parameter of austenite ($a_{o\gamma}$ in nm) at the mean steel composition (i.e. when $V_B=0$, V_B is measured volume fraction of transformation) can be calculated using equation determined by Dyson and Holmes [8]:

$$a_{o\gamma} [nm] = 0.3578 + \sum_{i=1}^n C_i x_i \quad (5)$$

where C_i are coefficients and x_i is the weight fraction of alloying element in phase γ , where $i = 1, 2, \dots, n$ represents alloying elements ($i = 1$ for carbon, $i > 1$ for substitutional solutes). The ferrite lattice parameter $a_{o\alpha}$ was determined by X-ray analysis and for ascertain it was calculated using following equation [9]:

$$a_{o\alpha} [nm] = 0.28664 + (3a_{Fe}^2)^{-1} \cdot [(a_{Fe} - 0.0279x_c^\alpha)^2 (a_{Fe} + 0.2496x_c^\alpha) - a_{Fe}^3] - 0.003x_{Si}^\alpha + 0.006x_{Mn}^\alpha + 0.007x_{Ni}^\alpha + 0.031x_{Mo}^\alpha + 0.005x_{Cr}^\alpha + 0.0096x_V^\alpha \quad (6)$$

The lattice parameter of pure ferrite was taken as $a_{Fe} = 0.28664$ nm [6]. The terms x_i^α represent the mole fraction of the species i in phase α . The carbon concentration of the residual austenite x_γ increases during bainitic transformation as a consequence of the increasing volume fraction of bainitic ferrite. Given that the excess carbon in the bainite ferrite partitions into the residual austenite and assuming that the final microstructure consists of only bainitic ferrite and residual austenite it is possible to estimate the carbon concentration x_γ , according to the following relationship derived from mass balance considerations [9]:

$$x_\gamma = \frac{2a_\alpha^3 \bar{x} (1 - x^\alpha) (1 - V_B) + a_{e\gamma}^3 (\bar{x} - x^\alpha) V_B}{2a_\alpha^3 (1 - x^\alpha) (1 - V_B) + a_{e\gamma}^3 (\bar{x} - x^\alpha) V_B} \quad (7)$$

The volume fraction of bainite (V_B) was determined as in [2,3,4,6,9,10] by dilatometry and additionally by means of a point counting method using light optical microscope and transmission electron microscopy (TEM) micrographs. The carbon content of the ferrite x^α , is a very small and is approximated by the A_1 $\alpha/(\alpha+\gamma)$ phase boundary of the Fe-C phase diagram (since of its smallness ignoring the effect of substitutional elements). The value of carbon in bainitic ferrite was taken to be $x^\alpha = 0.00139$ (0.03 wt.%) [6,11]. The factor $a_{e\gamma}$ is the lattice parameter of austenite after transformation, taking account of the carbon enrichment in austenite, when $V_B \neq 0$. The lattice parameters of bainitic ferrite and untransformed austenite at the reaction temperature, a_α and a_γ , can be calculated from the values of lattice parameters at ambient temperature (25°C, 298K) using the linear thermal expansion coefficients e_α and e_γ . The calculated carbon concentrations of residual austenite at the point where the formation of bainite

terminates will be next compared against the extrapolated T_0 , T_0' ⁱ and A_3' phase boundaries. The A_3' curve is the calculated paraequilibriumⁱⁱ ($\alpha+\gamma$)/ γ phase boundary indicating equilibrium between ferrite and austenite when the ratio of substitutional alloying elements to iron is constant everywhere. All the details of the determination of the carbon concentration of the residual austenite are described elsewhere [2-7]

MATERIAL, RESULTS AND DISCUSSION

The composition of the experimental steel investigated is given in Table 1.

Table 1. Chemical composition of the steel used in this study. All concentrations are given in wt. %

Steel	C	Mn	V	Si	P	S	Cr	Ni	Cu	Al
0.2C-1V-2Mn	0.17	2.00	1.17	0.28	0.031	0.020	0.10	0.07	0.10	0.03

The lattice parameters of ferrite and austenite and values of linear expansion coefficients are given in Table 2. The details for calculation of volume fraction of transformation are given in Table 3. The dilatometry results showed that the relative length change during the formation of bainite at selected temperatures increases as the isothermal transformation temperature decreases below the B_s , then the amount of bainite formed is dependent on the transformation temperature. Calculated and measured the carbon concentration of the residual austenite (x_γ) and carbon concentration at the selected interphase boundaries in 0.2C-1V-2Mn steel are given in Table 4.

ⁱ The T_0 temperature can be defined [6,9,12] such that stress free austenite and ferrite of the same composition (with respect to both the interstitial and the substitutional alloying elements) are in metastable equilibrium. Thus any displacive transformation involving a full supersaturation of carbon (i.e. bainitic ferrite would then inherit the carbon content of the parent austenite) can occur only below the appropriate T_0 temperature. The T_0' curve allows for 400J/mol of stored energy in the bainitic ferrite to take account of the strain energy due to the invariant-plane strain shape change that accompanies the growth of bainitic ferrite [6,12,13].

ⁱⁱ Hultgren [13], introduced the term “paraequilibrium” to describe the constrained equilibrium between two phases which are forced to have the same substitutional to iron atom ratio, but which achieve equilibrium with respect to carbon. Ferrite formation under conditions of paraequilibrium essentially implies that the substitutional lattice is configurationally frozen.

Table 2. Lattice parameters and linear expansion coefficients of ferrite and austenite in 0.2C-1V-2Mn steel

Steel	Lattice parameter of ferrite a_{α} nm	Lattice parameter of austenite a_{γ} nm	Ferrite ϵ_{α} $^{\circ}\text{C}^{-1}$, $\times 10^{-5}$	Austenite ϵ_{γ} $^{\circ}\text{C}^{-1}$, $\times 10^{-5}$
0.2C-1V-2Mn	0.28696 0.2868 *	0.35879	1.596	2.513

* The ferrite lattice parameter determined using a X-ray diffractometer

Table 3. Parameters for determination of volume fractions of bainite (V_B) in 0.2C-1V-2Mn steel

Transformation temperature, $^{\circ}\text{C}$	a_{γ} nm	a_{α} nm	$\Delta L/L^*$ $\times 10^{-3}$	V_B
370	0.36190	0.28854	3.63	0.86
400	0.36217	0.28867	3.12	0.74
433	0.36247	0.28883	2.49	0.59
463	0.36274	0.28896	1.69	0.40
490	0.36302	0.28911	1.03	0.24

* Values of the dimensional changes accompanying the transformation of austenite to bainitic ferrite measured at transformation temperature,
 V_B Measured volume fraction of transformation.

Table 4. Calculated and measured the carbon concentration of the residual austenite (x_{γ}) and carbon concentration at the selected interphase boundaries in 0.2C-1V-2Mn steel

Phase	Transformation temperature, $^{\circ}\text{C}$								
	700	600	560	490	463	433	400	370	320
	Carbon concentration at the phase boundaries, mole fraction								
A_3'	0.0093	0.0335	0.0454	0.0708	0.0801	0.0912	0.1049	0.1171	0.1347
$T_0(x_{T_0})$	0.0031	0.0092	0.012	0.0181	0.0203	0.0229	0.0265	0.0307	0.0365
$T_0'(x_{T_0}')$			0.0004	0.0076	0.0101	0.0132	0.0171	0.0207	0.0259
x_{γ}				0.0106	0.0131	0.0186	0.0265	0.0339	

The determined carbon concentrations of the residual austenite at the point where the formation of bainite ceases are compared with the T_0 , T_0' and A_3' phase boundaries for examined 0.2C-1V-2Mn steel in Fig. 1. The diagram was calculated using a model developed by Bhadeshia [2,6] based on the McLellan and Dunn quasi-chemical thermodynamic model [14]. The bainite and martensite reactions start temperatures B_S and M_S are also marked on that diagram.

Fig. 2 shows comparison of the measured and calculated carbon concentration of residual austenite at the termination of isothermal bainite formation in 0.2C-1V-2Mn steel but in case of bainitic reaction it is usually assumed that the points where dimensions cease to change does not represent full transformation.

Fig. 1. Calculated phase diagram with experimental data of carbon concentration of residual austenite at the termination of isothermal bainite formation for 0.2C-1V-2Mn steel (black circles represent experimental data)

Fig. 2. Comparison of the measured and calculated carbon concentration of residual austenite at the termination of isothermal bainite formation in 0.2C-1V-2Mn steel

Paraequilibrium is a constrained equilibrium in which only carbon atoms are free to partition between ferrite and austenite. The ratio of the substitutional solute concentration to the iron concentration is the same in ferrite and in austenite, the chemical potential of carbon is equal in the two phases [6,15]. The paraequilibrium phase boundary is chosen because no substitutional alloying element partitioning occurs during bainite formation.

If the hypothesis that paraequilibrium exists during all stages of transformation is correct, then the reaction should stop when the carbon concentration of the austenite is given by the A_3' curve. In presented experiment the reaction is found to stop when the average carbon concentration of the residual austenite is close to the T_0 curve than the A_3' boundary (black circles in Fig. 1). Note that the difference in carbon concentrations between the T_0' and A_3' curves at the chosen temperatures of bainitic reaction is very large, so the presented experiment is a sensitive indication of the failure of the transformation to reach paraequilibrium. The T_0' curve has a negative slope, so the austenite can tolerate more carbon before diffusionless transformation becomes impossible, as the transformation temperature is reduced.

The presented above results can be explained when it is assumed that bainitic ferrite grows without diffusion, but any excess of carbon is soon afterwards rejected into the residual austenite by diffusion [3,4,6,7,15]. This makes more difficult for subsequent bainitic ferrite to grow, when the austenite becomes stabilised by increased carbon concentration. The maximum extent to which the bainite reaction can proceed is therefore determined by the composition of the residual austenite. A stage where diffusionless growth becomes thermodynamically impossible and the formation of bainitic ferrite terminates is where the carbon concentration of the austenite reaches the T_0' curve. The incomplete reaction phenomenon supports the hypothesis that the growth of bainitic ferrite occurs without any diffusion with carbon being partitioned subsequently into the residual austenite. There is as yet no reasonable alternative explanation of this phenomenon.

The purpose of the light and TEM work was to determine the morphology of bainite and at which stage carbides are present during transformation. Figure 3 shows the light micrographs of microstructure at early stage of isothermal transformation at 490°C. Ferrite nucleation begins at austenite grain boundaries but autocatalysis* results in transformation developing preferentially toward the grain interiors rather than along the austenite grain boundaries. Saturation of the nucleation sites does not occur in this circumstance (Fig. 3 and 4). It is seen in Figures 3 and 4 that the morphology of bainitic ferrite is lath rather than plate. Bainitic ferrite laths form separately one from another. Several sheaves consisting of approximately parallel laths are seen in Figure 4. Large regions between bainitic sheaves correspond to the blocky austenite that transformed to martensite during quenching after the isothermal holding.

* The effect of autocatalysis results in the increase in number density of nucleation sites as the volume fraction of ferrite increases. The autocatalysis factor, familiar in martensitic theory describes the extent to which the formation of one plate stimulates others [6].

Fig. 3. Optical micrograph showing the mixture of bainitic ferrite and martensite after isothermal transformation at 490°C for 2 hours before quenching to room temperature. White etching phase, martensite is developed from austenite after quenching in water from temperature of isothermal reaction. Etched with 2% nital

Fig. 5 and 6 show the general morphology of the microstructure in 0.2C-1V-2Mn steel after austenitization at 1000°C and isothermal transformation at 490°C and 380°C. The morphology of the bainite is similar to low carbon lath martensite, where dislocated laths are separated by films of retained austenite. This structure, therefore, belongs to upper bainite assuming that upper bainite in this steel is a structure composed of carbides free bainitic laths with interlath retained austenite films [6,9].

Fig. 4. Optical micrograph showing the mixture of bainitic ferrite and martensite after isothermal transformation at 380°C for 1 hour before quenching to room temperature. White etching phase, martensite is developed from austenite after quenching in water from temperature of isothermal reaction. Etched with 2% nital

Fig. 5. Morphology of the microstructure in 0.2C-1V-2Mn steel after austenitization at 1000°C and isothermal transformation at 490°C for 2 hours. Thin foil

Fig. 6. Microstructure of 0.2C-1V-2Mn steel after austenitization at 1000°C and isothermal transformation at 380°C for 1 hour. Thin foil

After austenitization at 1000°C and at higher transformation temperature (490°C), bainite presents a morphology without carbides within the laths but with blocky retained austenite (Fig. 3 and 5).

Figure 6 shows TEM microstructure obtained by isothermal transformation to bainite at lower temperature at the early stage of transformation (380°C, 1 hour). In Figure 6 a bainitic packet is composed of bainitic ferrite and retained austenite. No carbide particles are observed at this stage. In early stages of transformation at 380°C the bainitic ferrite is carbide free.

The schematic stages of development of bainitic sheaf are shown in Fig. 7. As it is seen in Fig. 3 and 4 the bainite sheaves usually take the wedge shape. As indicated by many prior studies [2,3,6,9] new ferrite subunits are mostly nucleated near the tips of subunits on the sheaf, rather than at the broad sides. Thus, the different sides of the bainite sheaf may have different carbon concentration and different nucleation rates.

Figure 7 illustrates stages of development of bainitic sheaves. Suppose that the plate of bainite forms without diffusion, but any excess carbon is soon rejected into the residual austenite. In Fig. 7, 1—denotes subunits formed at early stage of transformation from austenite whose carbon concentration is initially identical to that of bulk alloy (region of upper bainite). 2 and 3—denotes subunits formed from enriched austenite as a consequence of carbon redistribution occurring after the growth event (probably region of lower bainite). The next plate of bainite then has to grow from carbon-enriched austenite (points denoted 2 and 3 in Fig. 8) because the carbon concentration of the residual austenite increases during bainitic transformation as a consequence of the increasing volume fraction of bainitic ferrite. Consequently any bainite which forms from enriched austenite will itself have a higher than bulk concentration of carbon (Fig. 7). The subunits 1, 2 and 3 correspond to the points 1, 2 and 3 on the phase diagram in Fig. 8. This leads to the possibility of the transformation beginning with the growth of upper bainite but with the enriched austenite then decomposing to lower bainite at the later stages of transformation. This explains why both upper and lower bainite sometimes can be found in the same microstructure.

Fig. 7. Schematic of stages of development of bainitic sheaves (t_1 , t_2 and t_3 – time of reaction)

Fig. 8 illustrates this situation in relation to the phase diagram. During isothermal transformation, a plate of bainite grows without diffusion and after this event excess of carbon is partitioned into the residual austenite. It means that the partitioning of carbon into the residual austenite occurs subsequent to transformation of subunits of bainitic ferrite. The next plate therefore has to grow from carbon-enriched austenite (it starts from the bulk carbon concentration $\bar{x}$ and passes through points 1, 2 and 3 in Fig. 8). The reaction is said to be incomplete, because the austenite has not achieved its equilibrium composition (given by the curve A_3') at the point the reaction stops.

Fig. 8. Schematic illustration of enrichment in the carbon concentration of untransformed austenite as the bainite reaction proceeds[6]

CONCLUSIONS

The paper presents an investigation of the austenite to bainite transformation in 0.2C-1V-2Mn steel using dilatometry backed by thermodynamic analysis. The following conclusions were reached:

1. The reaction terminates prematurely as the carbon content of the residual austenite reaches the T_0' curve well before the paraequilibrium carbon concentration is achieved (given by the extrapolated A_3' phase boundary) which indicates that the growth of bainitic ferrite is diffusionless. Therefore the carbon concentration of austenite can be estimated by the thermodynamics principles described here.
2. The extent of transformation to bainite in 0.2C-1V-2Mn steel, decreases when increasing the isothermal transformation temperature towards the bainite start temperature (B_S). This is because the austenite can only transform to bainite if its carbon concentration is less than a value $x_{T_0'}$ given by the T_0' curve.
3. The carbon concentration of the residual austenite increases during bainitic transformation as a consequence of the increasing volume fraction of bainitic ferrite.

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