Bainite - unanswered questions and why they matter

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Abstract: Atomic mechanisms of phase change are not just a curiosity, they help create innovative steels. The goal here is to consider what remains to be usefully explored in the context of bainite, and to set a formidable challenge.

1. THE QUESTIONS

a. Should we still justify research by the phrase *bainite is controversial*?

b. Why measure orientation relationships?

c. Continuity between Widmanstätten ferrite, bainite and martensite?

d. Why is bainite supersaturated with persistent carbon?

e. What is the big gap in theory?

2. SYNCHRONISED DANCE OF ATOMS OR CHAOS?

Tetrataenite (FeNi $P4/mmm$) has a high magnetic coercivity but only forms below 321 °C, so it occurs in some meteorites that cool from the red-hot state (> 1500 K) at 1-1000 K per million years [1]. But on Earth, artificial production of this permanent magnet is limited by the incredibly slow diffusion rate below its formation temperature. It has been estimated that nickel would execute one atomic jump in 10^4 years at 300 °C [2]. Bainite can grow at even lower temperatures (125 °C) where the diffusion distance of an iron atom is an inconceivable 10^{-17} m over the time scale of the experiment [3]. Even at 478 °C, the bainite reaction is 6000 times faster than the rate at which reconstructive ferrite can grow [4]. While there may be practical difficulties, there is in fact no theoretical lower-limit to the temperature at which bainite can form [5]; there is a 100 year experiment in progress to generate bainite at room temperature, scheduled to finish in the year 2104 [6]. The atomic mechanism of the $\gamma \rightarrow \alpha_{\mathrm{b}}$ cannot therefore involve the diffusion of atoms at the lattice points of the crystallographic unit cell of γ.

There are two main atomic mechanisms to consider, one involving "the complete disintegration [of the crystal] into bits and then *reconstruction* of its structure" [7–9], and the other where the change is achieved by a disciplined displacement of atoms into a new pattern. Although there has been much controversy in the past, it seems that the displacive mechanism is now accepted for bainite because key experimental observations cannot be reconciled with the reconstructive mode, for example, a recent paper from the school at KTH in Sweden [p.4547, 10].

However, consequences of the displacive mechanism are not sufficiently appreciated, leading to elementary errors. Remarkably, the large strain-energy due to the shape change in the transformed region,

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caused by the synchronised atomic-displacements [11] is neglected completely [12], in some cases to support a particular hypothesis [13]. In one publication, it is even argued incorrectly that transformation strain energy is "extremely small" when the volume fraction of bainite is small [14]; the strain energy *per unit volume of bainite* is independent of the volume fraction [15]. In fact, the invariant-plane strain shape deformation with its large shear component [11] dominates the shape and size of bainite, providing a way for steel manufacturers to produce grain sizes by phase transformation alone, on a length-scale much finer than can ever be achieved by thermomechanical processing [16, 17] or severe plastic deformation [pp.430-435, 18].

A second key feature of the transformation is that there is no partitioning of substitutional solutes, verified by numerous atomic scale observations across the transformation interface. Indeed, there is no segregation of any element, including carbon, at the semi-coherent interface (references in [17, 19]). And yet, there are articles being published to include arbitrary solute drag, or more generally, empirical treatments of the dissipation of free energy due to diffusion within the interface. It really is puzzling that none of the treatments that assume diffusion-controlled growth or its variants, take any account of the amazing structure where a bainite sheaf consists of small platelets, each of which grows only to a limited size [20, 21]. A displacive mechanism quantitatively estimates this structure on the basis of mechanical stabilisation due to the plastic relaxation of the shape deformation [22].

> So, here's a challenge: show that your theory explains all observations on bainite, not just those that are convenient. Show that it has been used independently in steel design, manufacture or critical experiments. I claim that the alternative 'models' fail in this test. Do at least attempt to prove me wrong!

A theory that recognises diffusionless $\gamma \rightarrow \alpha_b$ transformation and all its consequences [23, 24] has been applied generously to create innovative steels [17] and welding alloys [25, 26], and has been separately tested in both industry and academia. The associated software is freely available as source code [27]. A notable example is the ability to predict the chemical composition of austenite using the T_0 curve.

The manner in which atoms move during a change is important in determining structure. Most steels are designed to be weak ($\approx$ 300-500 MPa) because structural engineering requires numerous properties, including weldability, engineering rigidity, fire and earth-quake resistance, *etc.*, together with the ability to produce some 2 billion tonnes per annum at an affordable and sustainable price. The remarkable invention of microalloying and thermomechanical processing helped achieve the combination of properties, but the resulting finest ferrite grain-size associated with reconstructive transformation is about 1 μm [16]. In contrast, displacive transformation products grow as thin plates, which have a mean free-slip distance equal to twice the thickness of the plate – it therefore is possible to routinely generate grain sizes of 0.2 μm or even 0.02 μm by heat treatment alone.

Solutes do not partition during a displacive transformation so elements such as Mn, Cr, Mo, Si only affect bainite from a thermodynamic point of view, through $\Delta G^{\gamma\alpha}$, rather than from their mobilities. This of course is not the case for reconstructive transformations where the accumulation or depletion of solute at an advancing interface dramatically influences the rate. This is reflected in the much greater influence that these solutes have on the 'C'-curve that represents overall kinetics of reconstructive transformation in a time-temperature-transformation diagram, when compared against their influence on the corresponding curve for displacive products [Fig. 4.2, 18].

3. CARBON

Does the carbon partition during the growth of an individual bainite-plate, or is the transformation diffusionless, with partitioning occurring as an after-event? In the latter case, the $\gamma \to \alpha_b$ transformation can only occur if $T < T_0'$ [28]. If the carbon that is partitioned into γ remains in solution, then the maximum fraction $V_{V_{max}}^{\alpha_b}$ of bainite at any temperature is given by:

$$V_{V_{max}}^{\alpha_b} = \frac{\overline{x} - x_{\alpha_b}}{x_{T_0'} - x_{\alpha_b}} \qquad \text{with} \qquad x^\gamma = x_{T_0'} > \overline{x} \tag{1}$$

where $\overline{x}$ is the average concentration in the steel and the other terms are self-explanatory. Equation 1 and its foundational basis have been used to control the mechanical stability of the austenite retained at ambient temperature, in order to implement theory-based design and optimise mechanical properties without doing myriads of experiments [29–48].

Some of you may remember that at the first International Symposium on Steel Science held in 2007 at Kyoto, it was reported that time-resolved X-ray studies indicated that the austenite develops two lattice parameters just before transformation into bainite [49], possibly because of the old idea [50] that the austenite becomes unstable and splits into carbon-rich and carbon-depleted domains prior to transformation. Using greater resolution synchrotron X-rays, this has been shown to be untrue [51], independently verified using neutron diffraction [52].

However, it is expected and found [53] that the carbon in the *residual* austenite following transformation to bainite will not be homogeneously distributed for two reasons – the austenite is geometrically partitioned into films and blocks that would hinder homogenisation, and because the diffusion coefficient below B_S might be small. This is vividly illustrated in an optical micrograph of a partially transformed sample, where the sheaf of bainitic ferrite contains retained austenite in its perimeter, but martensite surrounds it [Fig. 3.5b, 17]. The blocky austenite can be mechanically unstable, leading to relatively large islands of brittle martensite that impairs toughness [29, 30]. In samples where the extent of the bainite reaction is large, the distribution of carbon is more uniform and consistent on average with the T_0 curve of the phase diagram. It is worth emphasising that the paraequilibrium $\alpha_b/\alpha_b + \gamma$ $Ae3'$ curve is far displaced to the right on a plot of temperature versus carbon concentration, so the reaction to bainitic ferrite definitely stops long before equilibrium or paraequilibrium is achieved. This is powerful evidence that the bainitic ferrite at first has the composition of the austenite from which it grows, though carbon may then partition from $\alpha_b \to \gamma$ or precipitate as a carbide within the supersaturated α_b if it autotempers.

The concentration x_C^γ is often compared against the negligible partitioning local equilibrium ('NP-LE') condition rather that T_0'. However, the calculated extent of the diffusion field of substitutional solutes is so small as to lose meaning, at the temperatures where bainite forms. It has been shown that the NP-LE mechanism does not exist [54] because there is a huge thermodynamic cost to creating steep concentration gradients [pp.204-207, 24]. Specifically, NP-LE calculations as currently implemented take no account of the gradient energy term, which if incorporated, would lead to long-range substitutional solute diffusion [p.116, 18]. But in any event, the NP-LE curve on a plot of T versus x_C cannot be used as a *limiting* condition, because the tie lines in the Fe-C-X phase diagram shift until equilibrium (as opposed to local equilibrium) is achieved. The T_0' curve, on the other hand, is a thermodynamic limit beyond which transformation without a composition change is truly impossible.

3.1. Carbon in α_b

There are extraordinary vestiges of diffusionless transformation to α_b. The carbon concentration in bainitic ferrite is far in excess of equilibrium, thought originally to reside in dislocations [53, 55]. Later work Pereloma and Caballero et al. showed that it is in fact in solid solution [56, 57], Figure 1a and

persists there even after 250 h at 200 °C, so it is not mobility that stops the carbon from partitioning into the residual-γ.
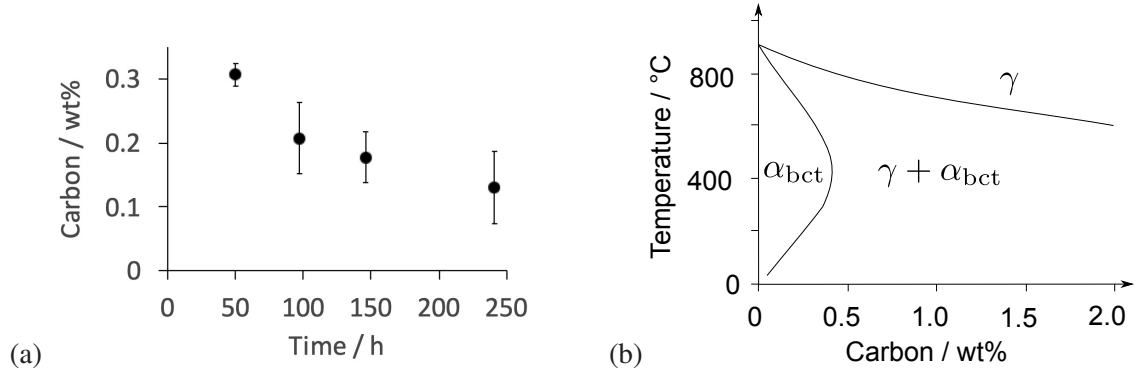

Figure 1: (a) Carbon dissolved in α_b during holding at 200 °C [58]. (b) Fe-C phase diagram for the equilibrium between body-centred tetragonal α and γ [59].

The reluctance of the carbon to partition from α_b^{bct} is because the Bain strain leaves it in just one of the three sub-lattices of the interstices, making the unit cell tetragonal; the residual tetragonality is verified experimentally [58, 60–65]. Equilibrium must therefore be considered is between α_b^{bct} and γ, not that between cubic-ferrite and γ. Figure 1b shows a calculated Fe-C diagram for the α^{bct}/γ equilibrium [59]; the solubility of carbon is increased significantly, explaining the large concentration that persists within α_b^{bct}.

Alternatively, carbon atoms have been suggested to associate with host vacancies in the α_b, thus compromising their mobility [66], but in fact the fraction of those atoms that pair with host vacancies is negligibly small [67], [pp.117-119, 24]. A third postulate that carbon is increasingly trapped in α_b at low temperatures [13] is not well-conceived because the driving force decreases as the austenite enriches, leading to a collapse towards equilibrium, unlike the T_0' concept which is a thermodynamic limit. In any case, if carbon is trapped by the moving α_b/γ interface then the transformation must be displacive because iron atoms move at many orders of magnitude slower pace.

4. PLATE SHAPE

There are only two reasons why a precipitate adopts a plate shape during solid-state transformation. If the interfacial energy while in contact with the parent phase is sufficiently anisotropic, then the diffusional growth-rate becomes direction dependent, especially when the driving force for transformation is small. This seems to represent the proeutectoid-cementite plates that form from γ in Fe-1.3C-13Mn wt% steel [68–70] and which are enriched in manganese concentration [71]. For this mechanism to operate, it would be necessary for some sort of orientation relationship to exist between the parent and product phases, to ensure a low interfacial-energy at the largest faces of the plates. It is interesting that relationships exist for θ plates but not for θ allotriomorphs that form at a similar temperature [72].

The second mechanism that generates a plate shape is displacive transformation because the strain energy per unit volume of transformed product is proportional to $cE_\gamma(s^2+\zeta^2)/\ell$ where E_γ is the Young's modulus, s and ζ are the respective shear and dilatational strains of the invariant-plane shape deformation, and c and ℓ represent the thickness and length, respectively, of the plate [15]. Therefore, a thin plate leads to a minimisation of strain energy, which is the term that dominates displacive transformations. The mechanism also ensures a defined level of coherency at the habit plane. Because of the disciplined movement of atoms, there *always* will exist a reproducible, irrational parent-product orientation relationship

[73–75]. Obviously, the iron to substitutional solute ratio is unchanged by transformation.

5.   NUCLEATION

Evidence-backed information on the nucleation of bainite is rare, but the following observations do fall into that category.

On the basis of the thermodynamic analysis of Widmanstätten ferrite (α_W) and bainite both evolve from the same nucleus by a mechanism in which the activation energy is a linear function of the driving force ΔG, with a required partitioning of carbon during the process. The nucleus then evolves into α_W or α_b depending on the driving force available for growth, once the surface-to-volume ratio no longer features significantly [23].

The linear dependence of the activation energy of nucleation on ΔG for bainite has its origins in a dislocation dissociation mechanism that was proposed by Olson and Cohen [76] in the context of martensite.

A combination of crystallography and three-dimensional observation has shown that α_b has a huge preference for nucleation at the faces of γ_1-γ_2 boundaries rather than at the edges where more than two grains meet [77]. It was found that nucleation triggered α_b into both γ_1 and γ_2 grains from the same site, with the bainite in the two grains having a similar orientation in space. This was deemed unlikely at γ grain-edges. The other interpretation could be that a mechanism based on the dissociation of (in this case interfacial) dislocations described above would be feasible at grain faces. This would also explain why many identically-oriented plates of bainite form at the same grain face.

Bainite in welding alloys nucleates intragranularly on particular inclusions present in the weld deposit [pp.251-285, 17]. Whether the mechanism is an enhanced driving force due to solute depletion around the inclusion, or as the inclusion absorbs manganese, or due to some sort of lattice matching, depends on the inclusion type. In another context, direct observations have shown that bainite can grow from allotriomorphic ferrite that is suitably oriented [78].

6.   CONTINUITY OF TRANSFORMATIONS

A comprehensive tabulation of the *observed* differences between Widmanstätten ferrite (α_W), α_b and martensite (α') is tabulated in [p.530, 24] where details can be found. Very briefly, these are all displacive transformations that exhibit an invariant-plane strain shape deformation, and the substitutional-to-iron atom ratio remains unchanged from the austenite. Martensite is diffusionless at all stages of its formation, α_b grows without diffusion but some of the excess carbon can partition into γ or precipitate as carbides depending on composition and conditions; it can form only below T_0, which is not a limitation for α_W that grows at a paraequilibrium, carbon diffusion-controlled rate in *steels*; it can also form in interstitial-free iron alloys [Chapter 7, Table 7.1, 24]. All this is quantitatively expressed [23, 27] in a generic manner. A previously unknown concept was predicted and validated, that not all three transformations are possible in all alloys.

The model that assumes continuity between α_W, α_b and α' with composition differences arising from changes in supersaturation [79] does not recognise that supersaturation changes at a constant temperature if the austenite composition alters, so at all temperatures, the system should collapse to paraequilibrium. Furthermore, the paraequilibrium concept itself is not defined for diffusional transformations [54]. That phase field simulations predict the plate shape of α_W through interfacial-instability in the carbon diffusion-field is wrong for many reasons, but the simple one is that α_W plates occur are common in carbon-free iron alloys [p.375, 24].

7. USEFUL AND SOME NOT-USEFUL EXPERIMENTS

a. The *crystallographic grain-size* was defined originally by Gourgues et al. [80] – it differs from the ordinary grain size because clusters of grains can have almost the same orientation in space. Therefore, the cleavage toughness deteriorates as the crystallographic grain size increases. More work of this kind in relating complex properties and structure in a quantitative framework would help design.

Routine measurements of the α_b/γ, carbide/α_b crystallographic orientation relationships, unless measured extremely accurately, are less useful. It is necessary to distinguish the Isaichev from the imaginary Bagaryatski orientation [72, 81, 82] and to define the precise irrationality of the α_b/γ instead of incorrectly referring to it as Kurdjumov-Sachs or Nishiyama-Wasserman. It follows that there always are 24 variants of bainite possible in an individual austenite grain.

b. The advent of crystallographic imaging has led to the misinterpretation of images. Such images are two-dimensional so they do not represent true size distributions, rather, areal or lineal intercepts, the mean of which defines just one size [83]. A true size distribution is difficult to measure, so it has to have an important, predefined purpose. It would be particularly useful to understand the geometrical partitioning of an austenite grain as the bainite transformation progresses. The evolution of the block and film morphologies of residual austenite that eventually is retained, can then be understood. The shape and size of austenite is really important in determining properties such as uniform ductility of TRIP-assisted and the Charpy toughness of strong bainitic-steels.

c. Is it necessary in practice to distinguish between stress- and strain-affected transformation? Stress-affected transformation is straightforward, where the loading is below the yield strength of austenite. Crystallographic variants of α_b that have shape deformations that comply with the applied stress are favoured over those that do not. All experiments on so-called strain-induced bainite actually have the simultaneous application of stress which drives the strain.

d. A huge problem in the design of bainitic steels is the chasm between mechanistic-theory and complex properties. It never has been possible to calculate the toughness, fatigue strength, elongation, creep strength, ... In mixtures of bainitic ferrite and carbon-rich retained austenite, we expect the austenite to improve such properties, even though during deformation it changes into high-carbon, untempered martensite. Such martensite is normally anathema to metallurgists. Is it not possible to produce a mixture of bainitic ferrite and low-carbon austenite? I predict that we will then be able to create the world's first bulk nanostructured steel with a toughness in excess of $150\,\mathrm{MPa\,m}^{1/2}$ and a strength of $\approx 2\,\mathrm{GPa}$, while resisting the pernicious effects of hydrogen.

8. SUMMARY

Steel is now a deep subject, not for the faint hearted. At the same time, it has no challengers as a structural material – witness its extraordinary growth and application. It is important, therefore, to focus on better understanding-based steel so that we reduce its production to quantities that are tolerable to the environment. I have written about a painless method to reduce consumption by 25% within four years, without jeopardising the quality of life that follows from steel [477-492, 18]. There is even a BBC radio programme introducing the idea [84].

I am not going to delve on the technical issues described in the main text, but suffice it to say that you are, of course, free to use or develop any theory that you feel is nearer the truth. But bear in mind the philosopher Sir Karl Raimund Popper who said 'a theory in the empirical sciences can never be proven, but it can be falsified'. It should be scrutinised.

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