Strength of mixtures of bainite and martensite

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Recently published experimental data demonstrate that the strength of mixed microstructures of tempered bainite and martensite can peak at an intermediate volume fraction of martensite. In the present work, a quantitative interpretation of these observations is achieved by modelling the mechanical properties of bainite and martensite in their tempered states. It is found that the peak in the curve of the strength as a function of the volume fraction of martensite can be attributed to two factors. When bainite forms it enriches the residual austenite with carbon, so that the strength of the subsequent martensite increases. In addition, during its deformation, the strength of the bainite is enhanced via plastic constraint by the surrounding stronger martensite. Taking these effects into account, it is possible to predict accurately both the trends and the absolute values of published experimental data on the strength of mixed microstructures. MST/1901

Introduction

Early indications[1] that mixed microstructures of bainite and martensite can often outperform the individual phases have been strongly reinforced by the more recent work of Tomita and Okabayashi.[2-7] The presence of bainite in a predominantly martensitic microstructure enhances both the strength and toughness relative to the single phase specimens, when the microstructures are in a tempered condition. For example, a maximum is observed for the strength of a mixture of lower bainite and martensite in an isothermally transformed high strength steel (Table 1) as a function of the volume fraction of lower bainite (Fig. 1). Tomita and Okabayashi suggest that the peak in the strength can be explained by the effective refinement of the austenite by the bainite sheaves, which in turn refines the martensite that forms subsequently. This strengthens the martensite, and the bainite itself is considered to be strengthened owing to the constraint to its deformation by the stronger martensite.

The purpose of the present work is to account quantitatively for the strength of mixed microstructures of bainite and martensite, including restraint effects and changes in martensite composition (and hence strength) as bainite forms.[8]

Strength of individual phases

The strength of martensite and bainite can be factorised into a number of intrinsic components[8,9]

$$\sigma = \sigma_{Fe} + \sum_i \sigma_{SS,i} + \sigma_C + K_L(\bar{L}_3)^{-1} + K_D \rho_D^{1/2} + K_P \Delta^{-1} \qquad (1)$$

where K_L, K_D, and K_P are constants, σ_{Fe} is the strength of pure annealed iron, $\sigma_{SS,i}$ is the solid solution strengthening due to substitutional solute i, σ_C is the solid solution strengthening due to carbon, $\bar{L}_3$ is a measure of the ferrite plate size, ρ_D is the dislocation density, and Δ is the distance between any carbide particles. The role of each of these components is now considered in detail.

IRON AND SUBSTITUTIONAL SOLUTES

The method used to estimate the strength of untempered martensite and bainite is a development of an earlier technique for steel welds,[9] where the strength is factorised into a number of components for which independent data are available in the literature.

Thus, pure bcc iron in a fully annealed condition makes an intrinsic contribution σ_{Fe}.[10-12] Substitutional solutes do not partition during the growth of either martensite or bainite, so their concentrations are fixed by the composition of the steel as a whole. Solid solution strengthening contributions $\sigma_{SS,i}$ can be estimated as a function of temperature and strain rate from published data.[9,13-19]

CARBON

Bainitic ferrite has only a small amount of carbon dissolved in interstitial solution, assumed to be 0·03 wt-%. Martensite on the other hand can have carbon concentrations well in excess of the average carbon concentration of the alloy $\bar{x}$, since the prior formation of bainite enriches the residual austenite according to the following relationship derived from mass balance considerations

$$x_\gamma = \frac{\bar{x} - V_b x_b}{1 - V_b} \qquad (2)$$

where x_γ is the carbon concentration in the residual austenite before its transformation to martensite, V_b is the volume fraction of bainitic ferrite, and x_b is the carbon that remains in the bainitic ferrite.

There is theoretical justification for assuming that the strength increment due to dissolved carbon should vary with the square root of the carbon concentration.[20] Data from Speich and Warlimont[21] are consistent with this

$$\sigma_{SS,C} = 1722 \cdot 5 x^{1/2} \qquad (3)$$

where strength is in MN m^{-2} and the concentration is in wt-%. This equation is used here to represent bainitic ferrite, but the data do not extend to the much larger concentrations appropriate for the martensite in the mixed microstructures. Thus, an earlier equation due to Winchell and Cohen[22] is used instead for martensite

$$\sigma_{SS,C} = 1171 \cdot 3 x^{1/3} \qquad (4)$$

with units as above. This avoids an overestimation of the strength of the martensite at high carbon concentrations, consistent with the experimental data of Winchell and Cohen.

DISLOCATIONS

The growth of both martensite and bainite in steels is accompanied by a change in the shape of the transformed region, a change which is described as an invariant-plane strain with a large shear component. When the transformation occurs at a temperature where the shape change cannot be accommodated elastically, the plastic deformation
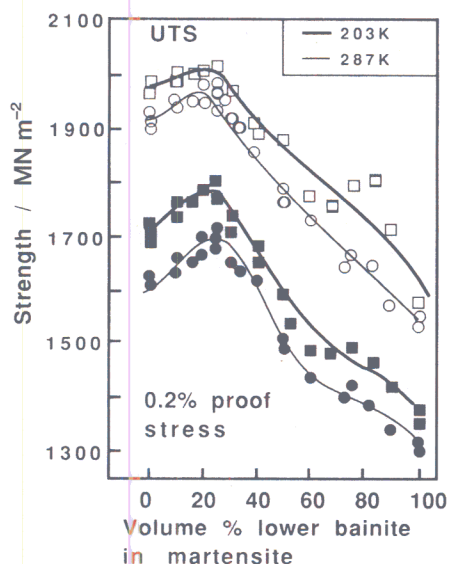
1 Variation in 0.2% proof stress as function of volume fraction of lower bainite in mixed, tempered microstructure of lower bainite and martensite of isothermally transformed high strength steel:[2] curves represent data collected at temperatures indicated on diagram

that is driven by the shape change causes the accumulation of dislocations in both the parent and product phases. Both the tendency for plastic accommodation and any recovery effects should depend largely on the transformation temperature. Therefore, it is not surprising that the dislocation density ρ_D of both martensite and bainite can be represented empirically as a function of temperature alone[23]

$$\log\{\rho_D\} = 9.2840 + \frac{6880.73}{T} - \frac{1\,780\,360}{T^2} \quad \ldots \quad (5)$$

where T is the transformation temperature in kelvin and ρ_D is stated in units of m^{-2}. The equation is based on experimental data over the temperature range 570–920 K. Although this covers most low alloy steels, in the present context the carbon concentration of the austenite that remains after partial transformation to bainite can be very high. Therefore, it is necessary in some cases to know the dislocation density of martensite which forms at temperatures below 570 K. The above equation cannot be extrapolated because of its empirical basis, but it is reasonable to assume that the dislocation density does not continue to increase indefinitely as the transformation temperature decreases. Instead, it should stabilise at some limiting value, since any reduction in dislocation density caused by dynamic recovery effects must become negligible at low temperatures. Thus, it is assumed here that ρ_D for $T < 520$ K is given by the value at 520 K (Fig. 2). The strengthening σ_ρ (MN m^{-2}) due to dislocations can be estimated to be[24]

$$\sigma_\rho = 0.38\mu b\rho_D^{1/2} \approx 7.34 \times 10^{-6}\rho_D^{1/2} \quad \ldots \ldots \quad (6)$$

where μ is the shear modulus and b is the magnitude of the Burgers vector.

LATH SIZE

The three-dimensional shape of martensite and bainite in low alloy steels is that of very fine plates or laths. However, in high strength steels of the type considered here, the shape is best described in terms of a plate morphology since isotropic sections are seldom observed. The strengthening σ_G due to the fine lath size then depends inversely on the mean value of the larger diameter of slip planes.[25,26]

Table 1 **Chemical composition of high strength steel used by Okabayashi and Tomita[2], wt-%**

C	Si	Mn	P	Cr	Mo	Ni
0.4	0.20	0.71	0.01	0.88	0.25	1.89

The aspect ratio (thickness/length) of the plates tends to be in the range 0.02–0.05, so that the mean slip plane diameter depends mainly on the thickness of the plates concerned. The thickness t is in turn given approximately by[8] $\bar{L}_3 = 2t$, where $\bar{L}_3$ is a mean lineal intercept measured at random orientations on random sections. Data such as these are not available in the literature, most results being reported as 'width' measurements, taken on random sections but not at random orientations – the measurements are usually at 90° to the traces of the habit planes. It is assumed here that these represent approximately $\bar{L}_3$ (μm). Thus, the strengthening due to the grain size of the plates σ_G (MN m^{-2}), for both martensite and bainite, is given by[8,25,26]

$$\sigma_G \approx 115(\bar{L}_3)^{-1} \quad \ldots \ldots \ldots \quad (7)$$

CEMENTITE PARTICLES

The strengthening σ_θ (MN m^{-2}) due to a uniform dispersal of spherical cementite particles is given by

$$\sigma_\theta \approx 0.52V_\theta\Delta^{-1} \quad \ldots \ldots \ldots \quad (8)$$

where Δ is the particle spacing and V_θ is the volume fraction of the cementite.

In the bainite that occurs in high strength steels, most of the carbon is partitioned into the residual austenite and remains in solution rather than precipitating as carbides. This is less true for lower bainite, but even for that phase much of the carbon is nevertheless partitioned into the austenite. In the present work it is assumed that cementite precipitation does not make a significant contribution to the strength, but that the carbon that is rejected into the austenite makes an important contribution via its effect on the solid solution strengthening of any martensite that subsequently forms as the austenite cools to ambient temperature or due to subzero treatment (as in the work of Tomita and Okabayashi).

Martensite composition and transformation temperature

Given that the excess carbon in the bainitic ferrite partitions into the residual austenite, which then transforms to martensite, it is possible to estimate the carbon concen-
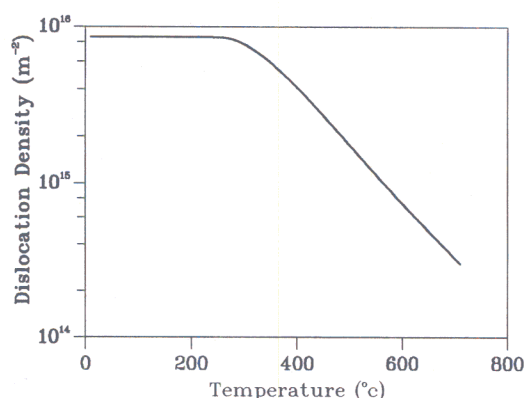
2 Assumed variation in dislocation density of martensite or of bainite as function of transformation start temperature
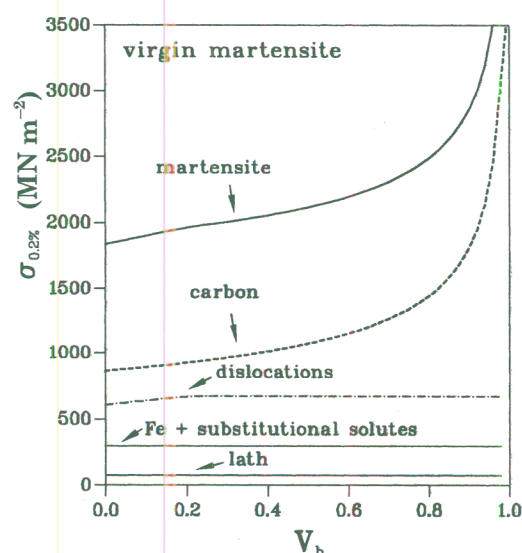
3 **Factorisation of strength of martensite into components (V_b volume fraction of bainitic ferrite)**

4 **Change in strength of martensite on tempering: note that carbon concentration of martensite increases as volume fraction of bainite increases**

tration according to the mass balance condition stated above (equation (2)). Following Steven and Haynes,[27] the martensite start temperature M_s of the residual austenite can be written as

$$M_s = M_s^\circ - 564(x_\gamma - \bar{x}) \quad \ldots \ldots \ldots \ldots (9)$$

where the concentrations are in wt-%, the temperatures in °C, and M_s° is the martensite start temperature of the austenite that has the average composition of the alloy. The M_s° temperature can be calculated using either the Steven and Haynes[27] equation or other methods.[28–30] Estimation of the martensite start temperature is important because it determines the dislocation density. The various contributions to the strength of martensite are illustrated in Fig. 3.

Effect of tempering

It is not possible reliably to use martensitic high strength alloys in their as quenched condition, without tempering heat treatments. Even when reasonable toughness might be achieved without tempering, there is a tendency for static failure to occur during service, as a result of hydrogen embrittlement. Thus, most high strength steels are used in the tempered state. The microstructures developed by Tomita and Okabayashi were tempered for 2 h at 473 K ($M_s^\circ = 586$ K).

Such a low temperature tempering heat treatment is not expected to affect significantly the strength of bainite. Bainitic and very low carbon martensitic microstructures are insensitive to heat treatment because of the small amount of carbon in solid solution. It is the precipitation of excess carbon which leads to large changes in strength during low temperature annealing of martensitic steels. Thus, the influence of the tempering heat treatment on the bainitic component of the microstructure is neglected here.

However, the same approximation is not valid for martensite which in the mixed microstructure is expected to contain a large concentration of carbon. The effect of tempering can be estimated using a recent model by Takahashi and Bhadeshia.[23] Assuming that hardness is linearly related to strength, that of tempered martensite σ_{MT} is given by

$$\sigma_{MT} = \sigma_M - \xi\{t\}(\sigma_M - \sigma_B) \quad \ldots \ldots \ldots (10)$$
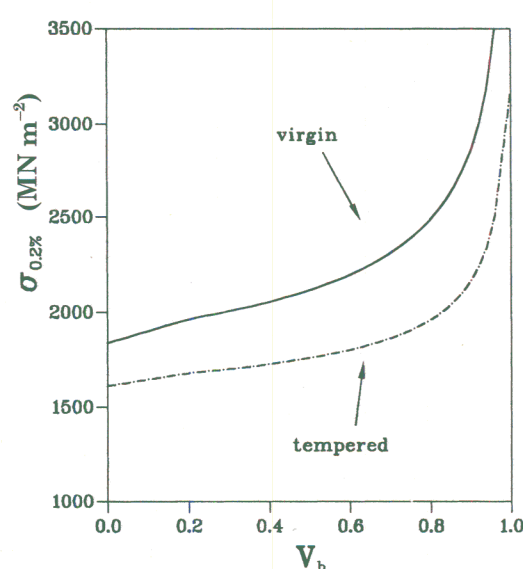
where σ_M is the strength of virgin martensite and σ_B that of martensite without any excess carbon (in the present case, equivalent to the strength of bainite). The Avrami function ξ is

$$\xi = 1 - \exp\{-kt^n\} \quad \ldots \ldots \ldots \ldots (11)$$

with $n = 0.62$ (Ref. 23), t is the tempering time (h), and k is given by

$$k = k_0 \exp\{-Q/RT\} \quad \ldots \ldots \ldots \ldots (12)$$

where $k_0 = 4.07 \times 10^4 \bar{X}^{0.635}$ ($\bar{X}$ is the atom fraction of carbon and the unit of k_0 is $h^{-0.62}$), $Q = 33\,598$ J mol^{-1}, R is the gas constant, and T is the absolute temperature.

Figure 4 illustrates the effect of tempering the martensite at a temperature of 473 K for 2 h.

Strength of mixed microstructure

It is well established in fracture mechanics that the yield strength is increased by plastic constraint. Hence, a weak brazing alloy can be used effectively to bond much stronger specimens provided that the thickness of the braze material is small enough to be constrained throughout by the surrounding stronger matrix. Indeed, the strength of the joint increases as the thickness of the braze layer decreases.[31]

Dispersions of bainite plates form in austenite which subsequently transforms to much stronger martensite. This is similar to a brazed joint in which the strong base material is connected to the soft braze. The deformation of the bainitic ferrite can therefore be expected to be constrained by the martensite. The effect can be modelled approximately by using experimental data[31–34] available from brazed joints in high strength steels. The brazing alloys used in making the joints were non-ferrous materials which are ordinarily rather weak. The data, in a normalised form, are summarised in Fig. 5. The strength is normalised with respect to the tensile strength of the pure unconstrained braze material, whereas the thickness is normalised relative to a value where the restraint effect vanishes.

The application of these data to the problem of a mixed microstructure of bainite and martensite relies on the assumption that the normalised thickness is equivalent to the volume fraction of bainite. This approximation should
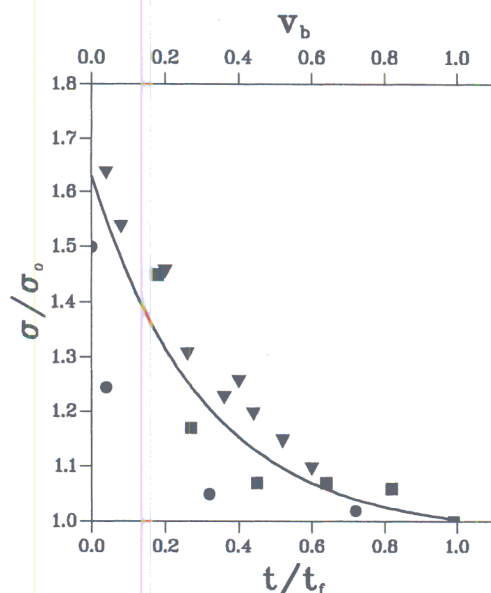
5   Plot of normalised strength of brazed joint versus normalised thickness of brazing material: data taken from Refs. 32–34

6   Illustration of contributions of bainite and martensite to overall strength of mixed microstructure

be taken as valid only at small volume fractions of bainite, but there are no difficulties at larger fractions since the constraint effect is expected to diminish rapidly as the volume fraction of bainite increases. Using this assumption, and the form of the normalised strength versus normalised thickness plot, the strength of constrained bainite may be represented by the equation

$$\sigma_B \approx \sigma'_B(0{\cdot}65\exp\{-3{\cdot}3V_b\} + 0{\cdot}98) \leqslant \sigma_M \quad . \quad . \quad . \quad (13)$$

where σ_B and σ'_B represent the strengths of constrained and unconstrained bainite respectively, σ_M is the strength of martensite, and V_b is the volume fraction of bainitic ferrite. Note that the strength of bainite is always restricted to be less than or equal to that of martensite. The above equation was used in the calculations to represent the strength of constrained bainite.

According to the formulation presented above, at low volume fractions (less than about 0·25) the strength of bainite is virtually equivalent to the strength of martensite (Fig. 6) whereas at all larger fractions the latter remains significantly above that of bainite considered alone. The strength of martensite continues to increase with the fraction of bainite, as the carbon concentration of the residual austenite from which it forms increases. This is an approximation; the very constraint which strengthens the bainite must make the martensite yield at a lower strength compared with a fully martensitic sample of the same composition.[35] However, this weakening of martensite by the presence of bainite is expected to be rather small in comparison with the very large effect of carbon on the strength of martensite. It is also worth noting that the strength of martensite does not continue to increase indefinitely as the carbon concentration increases (Fig. 6) but may saturate at about 2400 MN m^{-2} (Ref. 36). This again will have an insignificant effect on the calculations since the martensite approaches that strength level only when its volume fraction is very small.

Results and discussion

Tomita and Okabayashi have accumulated considerable data on the strength of mixed microstructures of bainite and martensite; all their data have been compared against
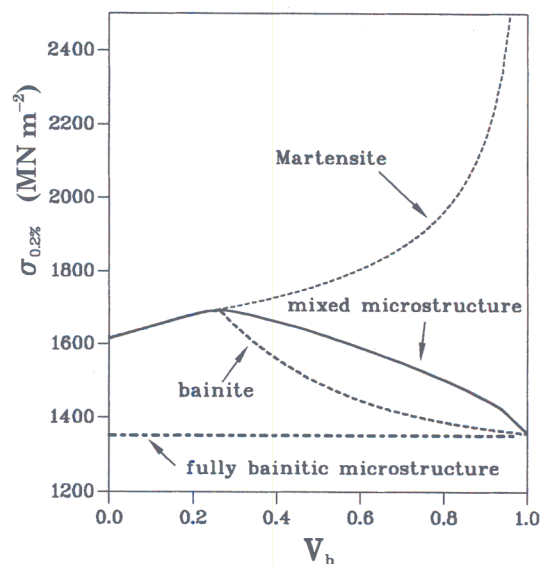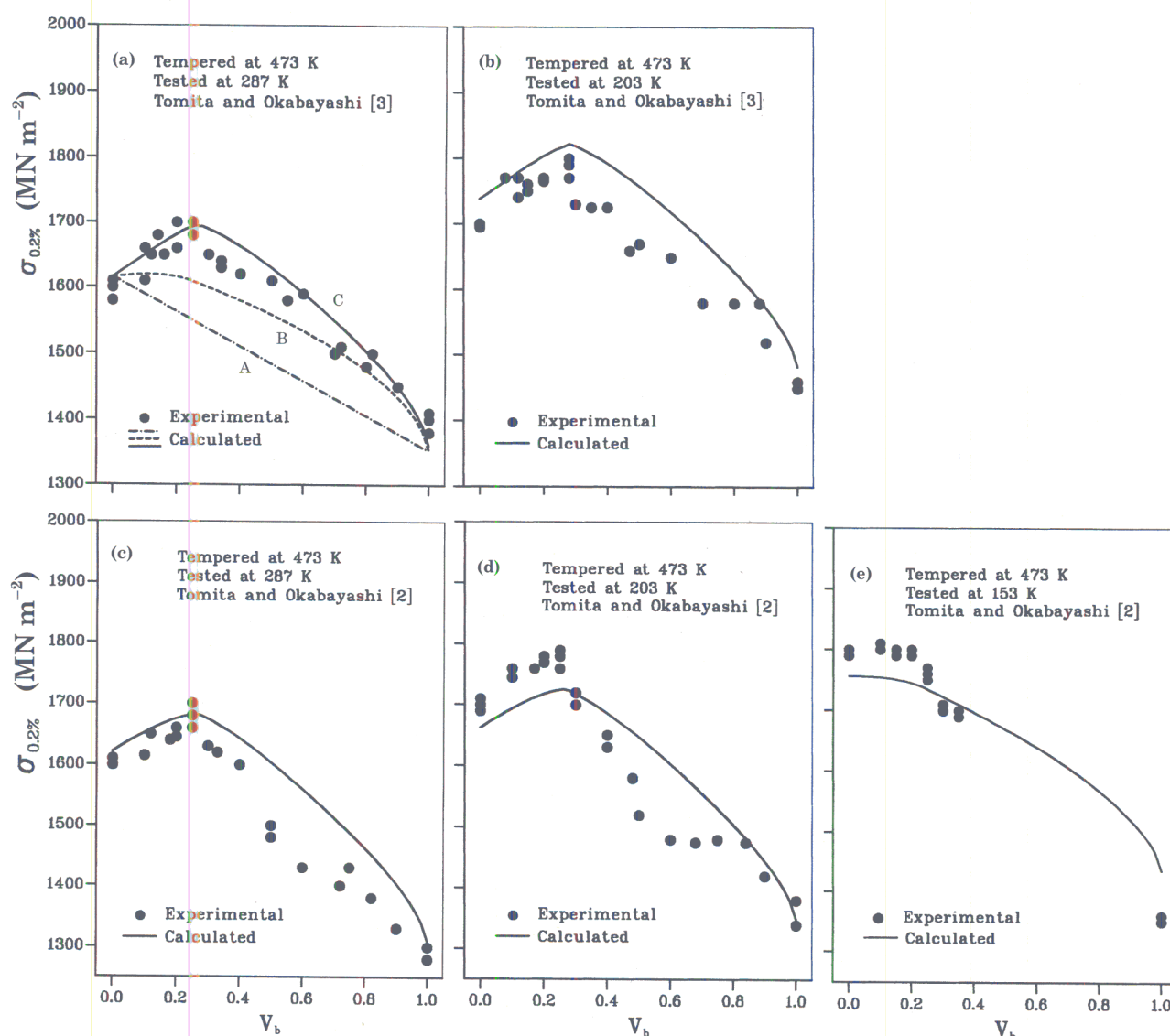
the above model. Figure 7a shows that the strengths of completely or partly martensitic specimens are accurately predicted, this being a general feature of the model as will be seen from further data presented below. Line A on Fig. 7a shows that a rule of mixtures cannot fully account for the variations observed. The agreement between calculation and experiment improves (curve B) as allowance is made for the change in the strength of martensite as carbon partitions into the austenite due to the formation of bainite. The consistency between experiment and theory becomes excellent when constraint effects are also included in the calculations (curve C).

Similar analyses are illustrated in Figs. 7b–7d, covering different steels and various test temperatures. The predictions of strength appear to deteriorate slightly as the test temperature is reduced (Figs. 7b, 7d). The variations in the strength of iron and the solid solution strengthening effects are included in the calculations; however, the temperature dependence of carbon, grain size, and dislocation strengthening are not considered owing to lack of appropriate data. Whether this accounts for the discrepancies is not clear because the detailed discrepancies do not appear to be systematic.

It is of interest that for the lowest temperature tests (Fig. 7e), neither experiment nor theory exhibits any significant peak in strength as a function of the volume fraction of bainite. The reason for this is that the strength of iron increases considerably as the temperature decreases, which means that the difference between the strength of martensite and bainite decreases, so that any constraint effect becomes less important.

Conclusions

It seems that the peak in the strength of mixed microstructures of bainite and martensite can be explained on the basis of two factors. The first is the increase in the strength of martensite as carbon is partitioned into the residual austenite during the formation of bainite. However, this does not fully account for experimental data. It is necessary to include a plastic constraint effect in which the strength of bainite is enhanced, particularly at low volume fractions of bainite, by the surrounding relatively rigid martensite.

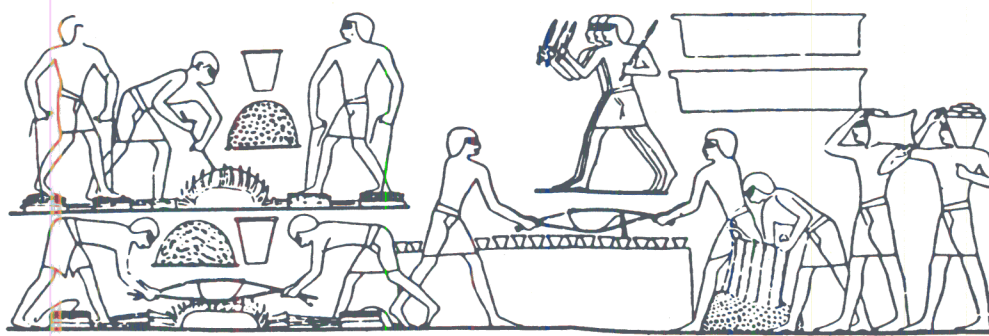**7   Comparison between experimental data[2,3] and theory**

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