# MICROSTRUCTURAL ENTROPY AND THE SCATTER IN TOUGHNESS

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## ABSTRACT

There is evidence that some of the scatter in toughness data from wrought and welded steels can be attributed to variations in microstructure (and hence mechanical properties). A simple method is developed to represent the degree of inhomogeneity in the primary microstructure of steels welds. The method involves the calculation of "microstructural entropy", using an equation which includes the volume fractions of all the phases deemed relevant, the equation being similar to one commonly used in deriving the configurational entropy of solid solutions. It is demonstrated, by comparison with experimental data, that the microstructural entropy can be correlated directly with the degree of scatter observed in toughness data. The limitations of the method are also discussed.

## INTRODUCTION

Fracture mechanics are widely applied in the design of engineering structures, but difficulties arise when repeated tests of the kind used in characterising toughness, on the same material, yield significantly different results. Such scatter in toughness is a common feature of relatively brittle materials such as ceramics, where it is a key factor limiting their wider application even when the average toughness may be acceptable. Steel users have become increasingly aware in recent years that scatter in toughness data can be of concern in wrought and welded steels [1,2]. Apart from the difficulties in adopting values for design purposes, the tests necessary for the characterisation of toughness as a material property are rather expensive, the number of experiments needed to establish confidence being larger for "unreliable" materials.

A major factor responsible for variations in toughness in welds is likely to be the inclusion population which consists mainly of large oxides originating from the slags used to control the weld pool stability and composition. The inclusions are neither uniform in size [3] nor are they uniformly distributed in the final weld [4]. There is also mounting evidence that variations in microstructure can also be an important factor in influencing scatter in toughness data [5-9]. Neville [10] noted that microstructural inhomogeneities such as hard pearlite islands, can lead to a significant variation in measured fracture toughness values during repeat tests on specimens of the same material. The purpose of the present work is to find a method of representing both scatter in toughness data,
and the degree of inomogeneity in microstructure, the latter being defined in a way which is physically significant and yet at the same time ameanable to alloy design techniques using phase transformation theory. The two quantities (scatter and heterogeneity) can the be related and tested.

## REPRESENTATION OF SCATTER

Due to a lack of other data, the present work is an analysis of scatter in toughness as assessed by impact testing. It is necessary to define a way of representing this scatter quantitatively, in a manner which normalises the data. The three most frequently used ways of rationalising variations in results from the toughness testing of weld metals are to take an average of the Charpy readings obtained at a given temperature (*e.g.* Ref. 11), measure the standard deviation [12], or plot the lowest Charpy readings obtained to focus attention on the lower ends of the scatter bands [13].

An alternative to this was suggested by [14] who proposed a scatter factor $SF$ to quantify any spread obtained in Charpy values, where

$$SF = \frac{MaximumEnergy - MinimumEnergy}{AverageEnergy} \times 100(\%) \tag{1}$$

This is essentially a definition of *range*; it has the disadvantage of giving excessive weight to the extreme values, taking account of all intermediate results only via the mean in the denominator, and also is in general an unpredictable function of test temperature.

None of the methods discussed above are completely suitable for alloy design purposes, in which the aim is to minimise scatter over the whole of the impact transition curve (the plot of impact energy versus test temperature). An idealised impact energy-temperature curve should be sigmoidal in shape, and the scatter of experimental data can in principle be measured as a deviation about a such a curve, obtained by best-fitting to the experimental data. The curve fitting can be carried out in three ways:

(a) the least squares method, which gives equal weight to all points;
(b) a weighted least squares method;
(c) a method in which the data are fitted to a logistic (log-related) curve.

In fact, the last method is most common for a sigmoidal, rather than plain curve, and appeared to be justified over an (unweighted) least squares analysis in that the residuals between the observed and fitted log/temperature scales (discussed below) were approximately the same at all temperatures, *i.e.* at steep and shallow gradients. A weighted least square analysis was not attempted since there was no clear way by which the weighting could be applied.

The sigmoidal curve has the form [15]:

$$ln\left\{\frac{E}{E_{US} - E}\right\} = \alpha + \beta T \tag{2}$$

where $E$ is the energy absorbed by the specimen during an impact test, $E_{US}$ is the upper shelf energy and $T$ is the test temperature. $\alpha$ and $\beta$ are experimentally determined constants.

A difficulty with fitting the sigmoidal curve to the experimental data is that the upper shelf energy needs to be defined. For the analysis, $E_{US}$ was taken to be an arbitrary 2% above the maximum recorded impact value. This treatment was found to be satisfactory, and is a fair assumption since the upper shelf energy is essentially independent of temperature over the range of interest [16]. $\alpha$ and $\beta$ are determined by plotting the intercept and gradient respectively of a graph of $ln\{\frac{E}{E_{US}-E}\}$ against temperature.

Regression analysis was carried out using the *General Linear Interactive Modelling* software of the Royal Statistical Society. The optimum values for $\alpha$ and $\beta$ occur when the scatter of a given set of data points around a trial curve is a minimum. The scatter may be evaluated by calculating a *deviance* of the data, which is equal to the sum of the squares of the deviations of the sample observations from the mean. this may be expressed algebraically as

$$\sum_{i=1}^{n}\left[ln\left\{\frac{E_i}{E_{US} - E_i}\right\} - \alpha - \beta T\right]^2.$$

The best values of $\alpha$ and $\beta$ are found, therefore , when this function is a minimum. However, in order to compare sets of data, it is necessary to consider the scale parameter

$$ScaleParameter = \frac{deviance}{\nu} \tag{3}$$

where $\nu$ is the number of degrees of freedom and illustrates the excess amount of data points available to be used in the regression analysis. It is defined as the number of data $n$ less the number of independent constraints on that set of data [17]. Since the equation has two unknown constants $\alpha$ and $\beta$, so $\nu = (n - 2)$.

The scale parameter allows for the fact that the deviance of a large set of data will necessarily be greater than that of a smaller set of equally scattered data. The attraction of this method is that it quantifies scatter irrespective of the actual shape of the curve or the absolute magnitudes of the data. It should be noted that this technique will give a false indication of the scatter associated with a given Charpy curve if only a few readings have been taken, and, irrespective of the proportions of various phases in the microstructure, if only three pairs of data are provided, the deviance will be zero! To guard against this, it is suggested that a minimum of some ten readings per curve should be taken.

## QUANTIFICATION OF HETEROGENEITY

Since it is believed that the scatter in Charpy data is amongst other factors, dependent on the inhomogeneity of weld microstructure, the degree of inhomogeneity needs to be quantified. This can be done by calculating the entropy $H$ of a given microstructure [18,19].

If $X$ is a random variable assuming the value $i$ with probability $p_i$, $i = 1, \ldots, n$, the entropy of $X$, as a logarithmic measure of the mean probability, is computed according to

$$H\{X\} = -\sum p_i ln\{p_i\}. \tag{4}$$

It should be noted that for $p_i = 1$, $H\{X\} = 0$. Conversely, the entropy is a maximum value $ln\{n\}$ when $p_1 = \ldots = p_n = \frac{1}{n}$.

The primary microstructure of most common welds can be taken as having three principal constituents: acicular, allotriomorphic and Widmanstätten ferrite. It is important to emphasise that although $\alpha_a$ and $\alpha_w$ have similar strengths [20], the weld metal microstructure cannot be treated as a two-phase microstructure (with $\alpha_a$ and $\alpha_w$ grouped together), since the *toughnesses* of the two phases are quite different. Therefore, the entropy of a given weld metal microstructure

$$H = -[V_\alpha ln\{V_\alpha\} + V_a ln\{V_a\} + V_w ln\{V_w\}] \tag{5}$$

where $V_\alpha$, $V_a$ and $V_w$ are the volume fractions of allotriomorphic, acicular and Widmanstätten ferrite respectively.

The entropy of the distribution quantifies the heterogeneity of the microstructure. $H$ will vary from zero for an homogeneous material to $ln\{3\}$ (*i.e.* 1.099) for a weld with equal volume fractions of the three phases. By multiplying by $1/ln\{3\}$, the heterogeneity of the three phase microstructure of a weld may be defined on a scale from zero to unity, *i.e.*

$$Het_3 = H \times 0.910 \tag{6}$$

Another scale of heterogeneity was also tested, to see if the primary and secondary regions of multipass welds could be treated similarly. Here, the secondary region is taken to comprise that part of the microstructure consisting of partially reaustenitised and significantly tempered regions [21].

It follows that the heterogeneity of the assumed two-phase microstructure is given by

$$Het_2 = \quad -[V_p ln\{V_p\} + V_s ln\{V_s\}] \times \frac{1}{ln\{2\}} \qquad (7)$$

where $V_p$ and $V_s$ are the volume fractions of the primary and secondary regions respectively.

## RESULTS

The initial work focused on an analysis of the toughness and as-deposited or primary microstructure of steel welds. Data were taken from Watson *et al.* [22], Fig. 1, and Bailey [23] for two pass submerged arc and triple arc submerged arc welds respectively, and results for the estimation of scatter, and calculation of heterogeneity are given in Tables 1 and 2. Although the data in [22] referred to one of the phases observed as proeutectoid ferrite, this is taken to be allotriomorphic ferrite in the present terminology.

Fig. 2 shows the relationship between the scatter observed in Charpy toughness values for the all-weld metal specimens and their microstructural heterogeneity.

Data for the calculation of $Het_2$ for the primary and reheated regions of multipass MMA low-alloy steel weld metals were taken from [13, 24]. The work by Taylor [13] is particularly convenient since the Charpy data had been presented in tabular form. The percentage of primary microstructure for Taylor's welds, which were in accordance with ISO-2560, could be estimated from a knowledge of the compositions and heat inputs of the welds [21]. It should be noted that the Charpy curves for W15SS and W15R [24] could not be included in this analysis, because the upper shelf energies for these welds were not evaluated. The analysis results are presented in Tables 3 and 4.

Fig. 3 shows calculated values for the scatter obtained in Charpy toughness experiments on multirun weld metal specimens, as a function of microstructural heterogeneity, treating the weld as a two-phase microstructure.

## DISCUSSION

It is evident from Fig. 2 that there is a strong relationship between the scale parameter, and microstructural heterogeneity for low-alloy steel all-weld metals. Consequently, a significant part of the observed scatter in weld metal Charpy results is attributable to the inhomogeneity of the microstructure, with larger scatter being associated empirically with more heterogeneous microstructures. This result can be compared with the common feature of fracture toughness experiments (*e.g.* [25]) where the positioning of the fatigue crack is found to be an important factor in COD testing of weldments.

The relatively poor correlation for the multipass welds (Fig. 3) highlights a limitation of this technique. The failure of the method for multirun welds might be attributed to the small range of microstructural entropy in the data analysed, so that the major differences in scatter are attributable to factors such as inclusions rather than microstructure. Secondly, the microstructural entropy term does not weight the phases involved in terms of their mechanical properties, but rather in terms of their volume

**Table 1** Estimation of scatter for all-weld specimens.

| Weld | Ref. | $E_{US}/J$ | Deviance | $\nu$ | Scale Parameter |
|---|---|---|---|---|---|
| AWO | 22 | 183 | 6.54 | 13 | 0.503 |
| FWO | 22 | 122 | 2.29 | 12 | 0.191 |
| AW5 | 22 | 134 | 6.96 | 11 | 0.632 |
| FW5 | 22 | 94 | 5.34 | 11 | 0.485 |
| W1 | 23 | 107 | 6.65 | 10 | 0.665 |
| W2 | 23 | 139 | 4.63 | 10 | 0.463 |
| W4 | 23 | 123 | 1.89 | 10 | 0.189 |

**Table 2** The heterogeneity for all-weld specimens.

| Weld | $V_\alpha$ | $V_a$ | $V_w$ | $H$ | $Het_3$ |
|---|---|---|---|---|---|
| AWO | 0.29 | 0.67 | 0.04 | 0.756 | 0.688 |
| FWO | 0.09 | 0.89 | 0.02 | 0.380 | 0.346 |
| AW5 | 0.50 | 0.47 | 0.03 | 0.788 | 0.717 |
| FW5 | 0.25 | 0.08 | 0.68 | 0.817 | 0.744 |
| W1 | 0.20 | 0.54 | 0.26 | 1.005 | 0.915 |
| W2 | 0.18 | 0.69 | 0.13 | 0.830 | 0.755 |
| W4 | 0.13 | 0.86 | 0.01 | 0.441 | 0.401 |

**Table 3** Estimation of scatter for multirun welds.

| Weld | Ref. | $E_{US}/J$ | Deviance | $\nu$ | Scale Parameter |
|---|---|---|---|---|---|
| W18SS | 24 | 184 | 1.10 | 8 | 0.138 |
| W18R | " | 181 | 2.07 | 8 | 0.259 |
| W19SS | " | 196 | 1.43 | 8 | 0.178 |
| W20SS | " | 200 | 3.41 | 8 | 0.426 |
| W20R | " | 199 | 2.86 | 8 | 0.358 |
| W22R | " | 197 | 4.60 | 8 | 0.575 |
| E7016 | 13 | 205 | 16.6 | 18 | 0.922 |
| E7016-1 | " | 221 | 18.2 | 15 | 1.212 |
| E7016-2 | " | 195 | 27.1 | 18 | 1.503 |
| E7016-3 | " | 192 | 10.5 | 18 | 0.582 |

**Table 4** Estimation of heterogeneity for multirun welds.

| Weld | $V_p$ | $V_s$ | $H$ | $Het_2$ |
|---|---|---|---|---|
| W18SS | 0.38 | 0.62 | 0.664 | 0.958 |
| W18R | 0.32 | 0.68 | 0.627 | 0.905 |
| W19SS | 0.33 | 0.67 | 0.634 | 0.915 |
| W20SS | 0.35 | 0.65 | 0.647 | 0.934 |
| W20R | 0.43 | 0.57 | 0.683 | 0.985 |
| W22R | 0.24 | 0.76 | 0.551 | 0.795 |
| E7016 | 0.30 | 0.70 | 0.611 | 0.881 |
| E7016-1 | 0.37 | 0.67 | 0.636 | 0.918 |
| E7016-2 | 0.42 | 0.58 | 0.680 | 0.981 |
| E7016-3 | 0.48 | 0.52 | 0.692 | 0.999 |

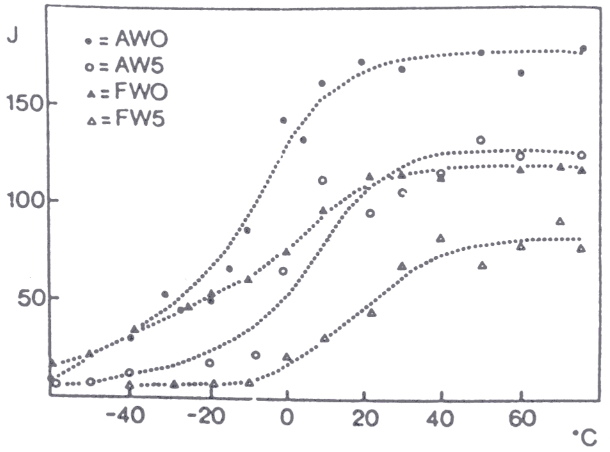

Fig. 1 - Charpy impact transition curves, used for one scatter analysis (data due to Watson *et al.*, 1981).

fractions. This is a significant weakness of the method, since it would predict heterogeneity in a multiphase system even when all the phases exhibit identical mechanical properties. For multirun welds, it is known that the strength of the reheated regions scales with that of the as-deposited regions [20], so that differences in mechanical properties in such welds should be less than expected intuitively.

## CONCLUSIONS

A method has been developed to represent the degree of heterogeneity of microstructure, based on a calculation of microstructural entropy. By relating this entropy to the degree of scatter observed in toughness measurements, it has been demonstrated that some of the scatter is attributable to variations in the microstructure. The work supports the conclusion [6] that weld deposits should be designed to be mechanically homogeneous.

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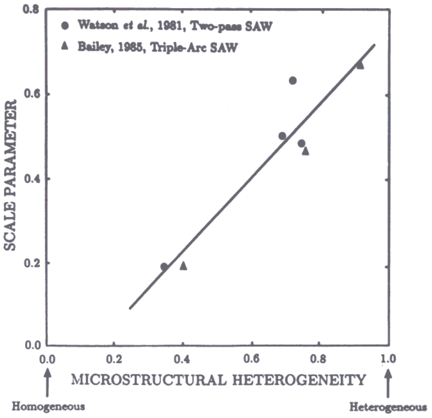

Fig. 2 - The relationship between microstructural heterogeneity and scatter, as measured by the scale parameter. Each point corresponds to a calculation using a complete Charpy impact transition curve. The correlation coefficient is 0.94.

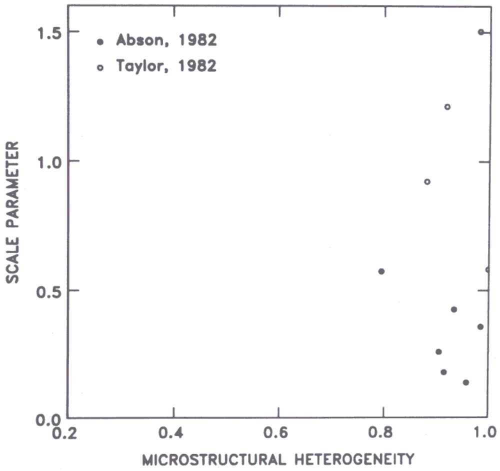

Fig. 3 - Microstructural heterogeneity versus scatter for the manual metal arc, multirun welds analysed.