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Superplasticity

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Abstract. Superplasticity is the phenomenon of extraordinary ductility exhibited by some alloys with extremely fine grain size, when deformed at elevated temperatures and in certain ranges of strain rate. To put the phenomenology on a proper basis, careful mechanical tests are necessary. These are divided into (i) primary creep tests, (ii) steady state deformation tests, and (iii) instability and fracture tests, all of which lead to identification of macroscopic parameters. At the same time, microstructural observations establish those characteristics that are pre-requisites for superplastic behaviour. Among the macroscopic characteristics to be explained by any theory is a proper form of the equation for the strain rate as a function of stress, grain size and temperature. It is commonly observed that the relationship between stress and strain rate at any temperature is a continuous one that has three distinct regions. The second region covers superplastic behaviour, and therefore receives maximum attention. Any satisfactory theory must also arrive at the dependence of the superplastic behaviour on the various microstructural characteristics. Theories presented so far for microstructural characteristics may be divided into two classes: (i) those that attempt to describe the macroscopic behaviour, and (ii) those that give atomic mechanisms for the processes leading to observable parameters. The former sometimes incorporate micromechanisms. The latter are broadly divided into those making use of dislocation creep, diffusional flow, grain boundary deformation and multimechanisms. The theories agree on the correct values of several parameters, but in matters that are of vital importance such as interphase grain boundary sliding or dislocation activity, there is violent disagreement. The various models are outlined bringing out their merits and faults. Work that must be done in the future is indicated.

Keywords. Superplasticity; ductility; stress and strain rate; grain boundary; dislocation creep; diffusional flow.

1. Introduction

Structural superplasticity is a high-homologous-temperature phenomenon exhibited by some metals and alloys which suffer extended or anomalous ductility under restricted circumstances. Elongations amounting to a few thousands per cent have been reported. When the conditions of deformation are changed, the same material does not possess the same ductility, and this has led to serious inquiries into the fundamental reasons for this behaviour. The technological off-shoot of this research, *viz.*, the development of many commercial alloys as well as forming processes, has not lagged behind. The aim of this paper is to summarize the observations made on superplastic materials and to identify their characteristics, to list the most important mechanisms identified as responsible for this behaviour, and to outline the more important models/theories presented so far. For recent reviews, see Edington *et al* (1976), Gifkins (1982), Hazzledine and Newbury (1976) and Padmanabhan and Davies (1980). Some points of controversy, which needs to be eliminated, are indicated.

2. Observations

The first systematic study on low-melting eutectics displaying superplasticity (Pearson 1934) is by now familiar. Observations on superplastic materials may be divided broadly into two classes: (i) those involving macroscopic aspects, from mechanical deformation studies; and (ii) those involving microscopic aspects, from microstructural observations.

2.1 *Macroscopic observations*

The variables are stress, strain, strain rate and temperature. However, a curve as shown in figure 1 is most common, showing the relationship between the flow stress σ and the strain rate $\dot{\varepsilon}$. This relationship can be mathematically written as

$$\sigma = A\dot{\varepsilon}^m, \tag{1}$$

where A is a constant and m is the strain rate sensitivity index (on a log-log plot, the slope of the curve). Three regions can be recognized which may conveniently be labelled I, II and III. Region II is the one of interest, where superplasticity is displayed. The peak values of m in this region coincide with maximum elongation, indicating a direct, if not linear, relation between them. In the superplastic region, m has values from 0·3 to 0·9. It may be mentioned here that all the three regions of figure 1 may not be found with a single experimental set-up.

The steady state strain rate in region II may be expressed in the familiar form of a diffusion-controlled process according to

$$\dot{\varepsilon} \propto \frac{Gb}{k_B T}(b/d)^p\,(\sigma/G)^n\,\mathrm{D}_0 \exp(-Q/RT), \tag{2}$$

where G is the shear modulus, b is the magnitude of the Burgers vector, and d the grain size. D_0 and Q denote a pre-factor and activation energy, respectively, while p and n are indices to be determined. Alternative equations to describe the behaviour of $\dot{\varepsilon}$ are also available in the literature. Their relative merits will be discussed in a later section.

The (thermal) activation energy in (2) may be determined by three different methods (Langdon 1982): (i) changing the temperature very rapidly during a test on a single

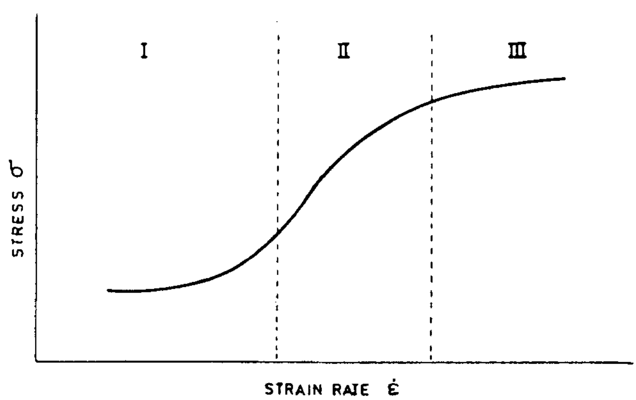

Figure 1. Flow stress plotted against strain rate.

specimen at constant stress; one finds

$$Q = \frac{R \ln (\dot{\varepsilon}_2 G_2^{n-1} T_2)\,(\dot{\varepsilon}_1 G_1^{n-1} T_1)}{[(T_2 - T_1)/(T_1 T_2)]}, \tag{3}$$

where $\dot{\varepsilon}_1$ and $\dot{\varepsilon}_2$ are the strain rates before and after the increase in temperature, and G_1 and G_2 the values of the shear modulus at the respective temperatures. (ii, iii) data from different specimens tested to steady state flow yield either

$$Q_\sigma = -R\left(\frac{\partial \ln \dot{\varepsilon} G^{n-1} T}{\partial(1/T)}\right)_{d,\sigma}, \tag{4}$$

or, in another version,

$$Q_{\dot{\varepsilon}} = R\left(\frac{\partial \ln (\sigma^n / G^{n-1} T}{\partial(1/T)}\right)_{d,\dot{\varepsilon}}. \tag{5}$$

We shall assume the equality of these two versions. Typical values of Q for the three regions suggest an activation energy for self-diffusion, Q_{SD}, in region I, an activation energy for grain boundary diffusion in region II, and an activation energy equal to or lower than Q_{SD} in region III.

The different activation energy values led to the belief that several mechanisms were operative. It must be noted here that $m(=1/n)$ varies with strain rate at different temperatures. The index m itself can be measured in several ways (Edington *et al* 1976). With the experimental values of n and Q, the grain size index p can be obtained by plotting $\dot{\varepsilon}$ *vs* the grain size at constant stress and temperature. A p value between 2 and 3 is typical of a number of superplastic materials in region II.

2.2 *Metallographic observations*

The microstructural study of superplastic materials includes surface observation through optical microscopy, replica electron microscopy and scanning electron microscopy, and internal structure determination through transmission electron microscopy. On a few specimens, fracture and cavitation have also been studied. Of great importance is the relative contribution of grain boundary sliding to the total strain. Microstructural observations made on specimens undergoing superplastic deformation show that: (i) there is no massive recrystallisation, (ii) there is grain rotation (both ways), (iii) a new surface is created, (iv) texture is generally reduced: (a) texture weakens continuously with strain; (b) texture weakens to a steady distinct lower level; (c) texture generally weakens, while retaining some components. (In some cases, new texture is introduced). (v) there is little dislocation activity, (vi) there is deformation near grain boundaries, (vii) grain boundary sliding takes place, (viii) there is diffusional flow in region I.

It is clear that any model proposed to explain superplastic behaviour must satisfactorily account for these observations. It must also lead to the proper constitutive equations to give the activation energy, strain rate sensitivity index and the grain size index. The general conclusions that may be drawn from the results of metallographic observations are listed in the next section.

3. Characteristics of superplastic materials

From the various observations made, the following general conclusions may be drawn (Sherby and Ruano 1982): (i) The superplastic material must have a fine grain size ($\leqslant 10\ \mu$m), which must remain so, (ii) the presence of a second phase is necessary to inhibit grain growth, (iii) the mechanical strength of the second phase must generally be of the same order as that of the matrix, (iv) the second phase, if harder, should be finely distributed within the matrix, (v) the grain boundaries between the matrix grains should be of the high angle kind, (vi) the grain boundaries should be mobile so as to reduce stress concentration at triple points, (vii) the grains should be equiaxed in order to enable a grain boundary to experience a shear stress, allowing grain boundary sliding to occur, (viii) the grain boundaries must resist tensile separation.

4. The role of grains

Noting that superplasticity is confined to materials with fine grains which remain fine-grained and equiaxed, one can conclude that the grain centres must move as if the material were deforming homogeneously (Hazzledine and Newbury 1976). Thus it is necessary that the grains slide past one another during deformation. Sherby and Ruano (1982) have considered two phenomenological equations

$$\dot{\varepsilon} = 10^9 \frac{D_{GB} b}{d^3} (\sigma/E)^2 \tag{6}$$

when grain boundary diffusion is thought to be the rate-controlling step, and

$$\dot{\varepsilon} = 10^8 \frac{D_{\text{latt}}}{d^2} (\sigma/E)^2, \tag{7}$$

when lattice diffusion is considered to be the rate-controlling step. Experimentally a plot of $(\dot{\varepsilon} d^3/bD_{GB})$ *vs* (σ/E) gives the correct exponent, namely, 2. Grain boundary sliding is therefore conceded to be the dominant mechanism.

Grain boundary sliding has been sought to be explained as brought about either by the motion of dislocations or by diffusion. In the dislocation model, crystal dislocations gliding to a grain boundary confine their subsequent motion to its vicinity. Special grain boundary dislocations whose Burgers vector is different and is related to a particular boundary may also move conservatively with the grain boundary. In this model, the strain-rate sensitivity m could be manipulated to be less than one, but then the grains would have to be elongated, which is not observed. On the other hand, the diffusion model considers the easy sliding of grains on smooth portions. If further sliding is opposed by irregularities, the rate of sliding is controlled by the movement or elimination of irregularities by diffusion. Such sliding could take place also at interphase boundaries. (These are classical notions and have been questioned by Padmanabhan and Davies 1980). However, since the strain rate is directly proportional to the applied stress, m will have a value of 1, which is at variance with experimental values. It would therefore appear that changes in the grain shape must also be taken into account.

4.1 *Grain strain*

Since structural continuity must be preserved, the grains must undergo some strain. This can be thought of in two different ways. Vacancy diffusion in the bulk can result in a change of shape, when the path of diffusion is fully across the grain. This Herring-Nabarro creep is given by the expression

$$\dot{\varepsilon} = (20\, D_{\mathrm{latt}}\Omega)/(k_B T d^2), \tag{8}$$

where Ω is the atomic volume. When the diffusion path is confined to the grain boundary region, one has Coble creep, given by

$$\dot{\varepsilon} = 50\, D_{\mathrm{GB}}\Omega\delta/(k_B T d^3). \tag{9}$$

Here δ is the width of the diffusion path. It may be noted that in both the above expressions, $m = 1$.

On the other hand, matter can be moved by conservative and non-conservative motion of dislocations. Nabarro (1967) has given two expressions, the first being a power dependence $\dot{\varepsilon} \propto \sigma^3$; the second for pipe-diffusion along the cores of dislocations, leads to $\dot{\varepsilon} \propto \sigma^5$. It may be mentioned that the grain size does not figure in these expressions. Friedel creep envisages a pile-up of dislocations against a grain boundary, from which the leading dislocation climbs into the grain boundary. The process is controlled by the climb rate of dislocations. This has been utilized to yield a relation $\dot{\varepsilon} \propto \sigma^2$. However, it is clear that grain elongation is unavoidable, and that as the grains elongate, the process must be exhausted. There is nothing in this picture to indicate the rotation of the grains. Moreover, no dislocation pile-ups have been observed in superplastic alloys.

4.2 *Interaction between grain strain and grain boundary sliding*

It is commonly believed that neither of the above can take place without the other, as they must together produce strain rates compatible with one another. The amount of strain produced by grain sliding is usually measured by surface markers, even though there is considerable controversy about the measurement. However experimentally the value $(\varepsilon_{\mathrm{GBS}}/\varepsilon_{\mathrm{tot}})$ varies between wide limits. It may be pointed out here that such measurements have been made at a strain of about 0·2, which is not typical of superplastic deformation. Stevens (1971) puts an upper limit of 0·62 to this ratio. Hazzledine and Newbury (1976) point out that at any instant the strain *rate* due to sliding is of the same order as the *rate* of strain of the grain.

Nevertheless, the idea has persisted that the strain due to grain boundary sliding must necessarily be less than the total strain. Consequently, an overwhelming majority of the mechanisms suggested invoke some process or the other to accommodate grain boundary sliding. It is against this background that different mechanisms are outlined in the next section. It is instructive to reproduce the figure from Edward and Ashby (1979) illustrating the different accommodation processes (figure 2).

5. Atomic mechanisms for superplasticity

It has been pointed out earlier that mechanisms employing diffusion creep or slip are unable to account for many of the observations satisfactorily, in particular for the large

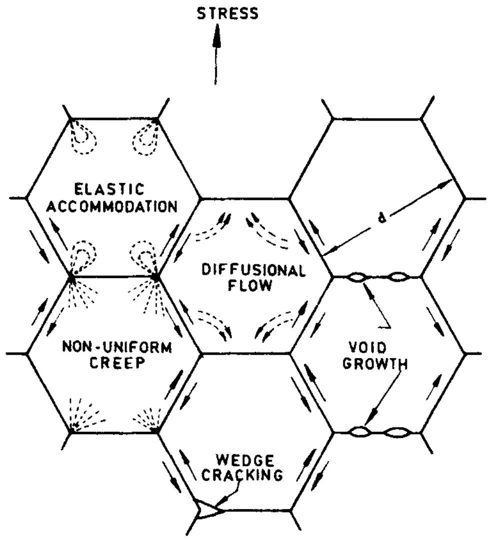

Figure 2. Consequences of grain boundary sliding (After Edward and Ashby 1974).

amount of grain boundary sliding. In this section, some more mechanisms will be listed where grain boundary sliding (GBS) is considered along with some accommodating processes.

5.1 *GBS accommodated by dislocation motion*

Ball and Hutchison (1968) put forward the idea that several groups of grains slide at once. When there is an unfavourably oriented grain, a stress concentration results which can be relieved by dislocation motion. The dislocations, which are necessarily in a pile-up, prevent further sliding until the lead dislocation climbs. Mukherjee (1971) contended that the grains move individually. Again a pile-up is invoked because dislocations are produced by irregularities in the grain boundaries. This itself is a major difficulty since, as mentioned earlier, no pile-ups have been seen. Moreover, the same dislocation motion will be rate controlling in region III too, and it would be impossible to distinguish between the two.

5.2 *GBS and diffusion creep*

Ashby and Verral (1973) proposed a grain switching event as central to their model. A group of four grains reacts to a tensile stress, sliding past each other and changing their shape to maintain continuity. These switching events occur randomly throughout the specimen with various clusters in different stages of the process. The accommodation strain is accounted for by bulk and boundary diffusion. While the model, which considers the topological aspects of the sliding process, satisfactorily explains most of the observations, there are large variations in the predicted values of parameters.

Hazzledine and Newbury (1976) have used the concept of grain emergence: as a group of adjacent grains slide, a grain from the layer below emerges at a location where a fissure would otherwise open up. Grain boundary migration occurs at the same time to restore the dihedral angles, resulting in the rounding off of the emergent grain and the curvature of all other grain boundaries. Having conceded that the phenomenon is too complicated for analysis, Hazzledine and Newbury proceed to give a viscoelastic model of superplasticity with several dashpot elements representing Nabarro, Coble and Friedel creep, as well as grain boundary and interphase sliding. The general conclusion is that a single comprehensive mechanism is not possible.

5.3 *GBS and core-mantle theory*

Gifkins (1976) proposed for each grain a non-deforming core surrounded by a mantle in which flow occurs. While the core contains statistically stored dislocations, the mantle, which is a few per cent of the grain diameter, contains geometrically necessary dislocations. The grain boundary dislocations piled-up against a triple edge are responsible by their movement for grain boundary sliding. The stress concentration of the pile-up is regarded as that of a freely slipping crack. By climb and glide the lead dislocations accommodate grain boundary sliding. Gifkins arrived at an appropriate equation to account for the observed parameters. Since the widths of the core and mantle are adjustable parameters, the experimental verification has been criticised as "an exercise in curve fitting". Further, no single mechanism has been identified as responsible for superplasticity.

Arieli and Mukherjee (1980) have also criticised this model on the ground that the deformation will slow down in time and eventually come to a stop, since many dislocations will be lost by annihilation. Also the stress concentration has been calculated by the Eshelby, Frank and Nabarro method, which does not apply to climb or any other deformation process.

Arieli and Mukherjee themselves employed a modified mantle behaviour. The small number of pre-existing dislocations are attracted to the boundary under the action of the applied stress and their own line-tension. The individual dislocations climb the short distance to the boundary and, in the process, create new dislocations by the Bardeen-Herring mechanism. At the boundary, the dislocations are annihilated. The eventual stress concentration created by the dislocations climbing into the boundary are relaxed by grain boundary diffusion. It may be noted here that there has been no experimental evidence for the Bardeen-Herring mechanism.

5.4 *Micromultiplicity*

As the grain boundary network has alternate sets of shear paths, the deformation continues even if grain boundary sliding is obstructed locally. In other words, the local stress conditions would alternatively enhance or decrease the importance of the obstacles. A number of processes, mentioned above, could be appropriately combined to give the total strain rate. It is obvious that this is also merely a curve-fitting exercise since the arbitrary constants of the constitutive equations can be adjusted appropriately.

5.5 *Unaccommodated GBS*

There has been considerable controversy as to whether grain boundary sliding is independent of diffusional strain. The work of Raj and Ashby (1972), Speight (1975) and Beere (1976) has shown that specimen strain can only develop when both sliding and diffusion can operate simultaneously. Therefore the deformation can be attributed to either sliding or diffusion, and can be described as diffusion-accommodated sliding or sliding-accommodated diffusion; in no sense can they contribute independently (Burton 1977).

Accordingly, Padmanabhan (1977) regards the upper limit of 60–65 % placed on the contribution from sliding to be a consequence of the assumption that both diffusion and sliding are present as separate steps. It is also held that the ideas concerning sliding in coarse-grained materials were not necessarily relevant to sliding in ultrafine-grained alloys, and that superplastic deformation is a result of pure grain boundary sliding of non-deformation grains that require no accommodation. Starting from the fundamental rate equation one arrives at a constitute equation

$$\dot{\varepsilon} = C\frac{\delta}{d^2}\sigma^n \exp(-Q_0/RT), \tag{10}$$

where C is a constant and δ is the jump distance in the grain boundary region. The stress exponent n may be recognized as the inverse of the strain-rate sensitivity index, m, and is shown to be a function of stress, grain size and temperature. Excellent agreement with experimental results is achieved, as is a demonstration of the transition to the region III in which climb-controlled motion of dislocations occurs.

6. The activation energy for superplasticity

The activation energy Q has been introduced in (2). It was mentioned there that different values are obtained for Q in the three regions of figure 1. This is done by treating the sigmoidal curve as a combination of three linear curves. The assumption of the equality of Q_σ and $Q_{\dot{\varepsilon}}$ ((4) and (5)) has also been mentioned. Padmanabhan (1981) has objected to the procedure on the grounds that (2) is empirical, that for each range a constant but different n value is assumed, and that within each range the activation energy is assumed to be independent of the temperature and the stress level of its evaluation. He has pointed out that the evaluation of the activation energy from an equation that has no physical basis is bound to lead to erroneous results, and that since n is continuously variable, there is no justification for treating this as a constant. At the junctions of region II with regions I and III the n value becomes ambiguous. Topologically and microstructurally regions I and II are not different. Only region III indicates a change of mechanism. Another serious objection is the treatment of an expression containing a T-dependent pre-factor as a true Arrhenius form. Padmanabhan (1981) has derived expressions for Q_σ and $Q_{\dot{\varepsilon}}$ (the values corresponding to the shear mode) and has pointed out that the best estimates of the real activation energy are only attained when data pertaining to the range in which m is large (~ 1) are used. It is regrettable that many of the leading workers in the field have not recognized the force of these arguments.

7. The role of dislocations in superplasticity

Even though many of the models invoke dislocations to explain superplasticity, there has been no direct experimental evidence to support dislocation activity. However, Edington *et al* (1976) have argued, taking Nicholson's (1972) experiments as an example, that dislocations might have been active or that they might have been obstructed by precipitates. Padmanabhan (1980) has argued that this was unlikely. He has further held that any dislocation seen was the residue of the initial configuration or the existence of local stress concentrations or a favourable strain rate at which dislocation motion commences. *In situ* experiments have also failed to reveal substantial quantities of dislocation.

Melton and Edington (1973) have maintained that since (i) statistically significant differences in the angular distance of the dislocations from their nearest possible glide planes were found as a function of the deformation rate; (ii) the Burgers vector distribution changed as a function of strain rate; (iii) increasing the initial density by cold working prior to superplastic deformation did not produce any detectable differences in the Burgers vectors and densities after superplastic deformation; (iv) there is an equilibrium dislocation density typical of superplastic flow and (v) dislocations are also observed at strain rates an order of magnitude less than that corresponding to m_{max}, one must conclude that crystallographic slip plays a small but significant role in superplasticity. Padmanabhan (1980) has countered these arguments by citing experimental results accepted by Melton and Edington themselves. It therefore appears that the available experimental evidence could support either point of view. Lücke (1974) has pointed out that any sequence of deformation processes leading to a given strain tensor will give the same texture. Hence the operating mechanisms cannot be deduced solely from texture results. TEM has provided no evidence for dislocation activity.

8. Future work

To set at rest the controversy regarding the mechanism of superplasticity, work is required along the lines indicated below: (i) a better understanding of the structure of grain boundaries and interphase boundaries, (ii) a correct analytical form for the flow stress as a function of strain rate, temperature and grain size, (iii) unequivocal experimental evidence to support or discount dislocation activity, (iv) an understanding of the role of the stacking fault energy, (v) an unambiguous interpretation of experimental curves such as the double sigmoidal plot of figure 1.

It is to be hoped that there will be a happier conclusion to superplasticity than in the case of work hardening, in which field the workers have apparently agreed not to agree.

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