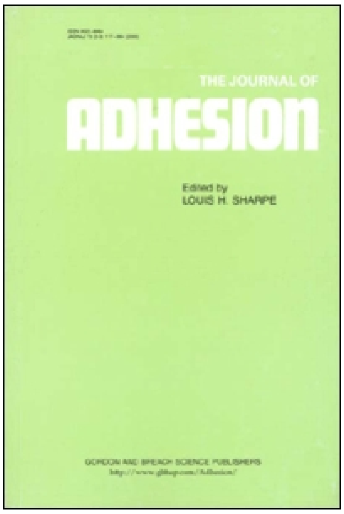

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The Effect of Interfaces on the Behavior of *In-Situ* Metallic Composites†

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The morphology and properties of an *in-situ* metallic composite depend on the nature of the interfaces separating its constituent phases. In this paper, four models for these interfaces are considered: coherent, semi-coherent, cusp-oriented and noncoherent. The consequences of each model with respect to composite morphology, load transfer, morphological stability at elevated temperatures and mechanical behavior are discussed.

I INTRODUCTION

In-situ metallic composites with aligned microstructures have been produced by a variety of techniques. These techniques include the unidirectional solidification of eutectic alloys,[1] the unidirectional transformation of eutectoid alloys,[2] and magnetic aging[3] or stress aging[4] of alloys with coherent precipitates.

The unidirectional solidification of a eutectic alloy to produce an aligned composite structure involves cooling the alloy from the melt in a mold arrangement where heat flows primarily in one direction. During solidification, the solid–liquid interface moves parallel but opposite to the direction of heat flow. For alloys where the constituent phases have low entropies of melting, as is the case in many metallic systems, unidirectional solidifica-

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tion produces a composite microstructure with fibers or lamellae aligned parallel to the growth direction.[5] Alloys where the volume fraction of one phase is less than $\sim$0.3 usually from fiber composites with the minor constituent as the dispersed fiber phase.[6]

Aligned composite structures can be produced from eutectoid alloys by a technique similar to that described in the preceding paragraph. The alloy is cooled from a temperature above the eutectoid temperature by the unidirectional extraction of heat. The transformation proceeds behind a planar interface that moves parallel but opposite to the direction of heat flow. Slower transformation rates are required for the eutectoid reaction than for the eutectic reaction because the former is controlled by diffusion rates in the solid state. Only aligned lamellar eutectoid microstructures have been produced.

In magnetic aging and stress aging, an alloy containing coherent precipitates is aged while in a magnetic field or in a state of uniaxial stress an aligned composite structure is formed as the consequence of coarsening processes.

There are important differences between the "*in-situ*" composites discussed in this paper and the "fabricated" composites considered by Metcalfe and Klein.[7] The fabricated composites are normally formed from dissimilar materials by processes involving relatively low temperatures and short times so that a state of chemical equilibrium is not obtained. Their properties are strongly influenced by the degree of chemical reaction at matrix–fiber interfaces. In applications involving exposure to temperatures high enough for additional reaction to occur, the properties of this type of composite may change with time. In contrast, the *in-situ* composites are formed by processes such as solidification, precipitation or coarsening at temperatures where chemical equilibrium can be obtained. These composites are better suited for applications at elevated temperatures than the fabricated composites; however their properties may also change after long time exposure due to processes decreasing interfacial energy or elastic strain energy.

Another important difference between the fabricated and *in-situ* composites is related to scale. Table I gives fiber diameters and fiber spacings for typical fabricated and *in-situ* composites. The comparison suggests that different approaches may be required to understand the nature of interfacial stress relaxation in fabricated composites than in *in-situ* composites. In the former, interfacial stress relaxation has been treated as a bulk yielding or interface sliding process.[8] In the latter, it may be better understood in terms of a more detailed dislocation mechanism (Section 3.2.1).

Of the various types of *in-situ* composites that have been mentioned, only the unidirectionally solidified eutectics have been seriously considered for structural applications. In what follows, the types of interfaces that occur

TABLE I
Typical fiber diameters and spacing for *in-situ* and fabricated composites

Composite	Vol. Pct.	$D(\mu)$	$\lambda(\mu)$
BORSIC®-Al	50	107	145
THORNEL 50®-Al	50	8	11
Al_3Ni–Al[a] eutectic	10	0.5	1.6

[a] Solidified at 10 cm/hr.

in this kind of composite and their role in determining its properties will be discussed.

II TYPES OF INTERFACES

The most important microstructural features in unidirectionally solidified eutectics are the grains, phases and cells. Grain boundaries and phase boundaries are true interfaces in the sense that they join dissimilar homogeneous regions. The former join regions differing in crystal orientation; the latter, regions differing in crystal structure and composition. Cell boundaries are not true interfaces by this definition. They are better described as growth faults and will not be included in this discussion.

2.1 Grain boundaries

The grain structure in unidirectionally solidified eutectic ingots[1,9] is similar to that observed in other unidirectionally solidified alloys.[10] Nucleation at a chill surface produces many small randomly oriented grains. In subsequent growth from the chill a columnar grain structure develops with the long dimension of each columnar grain lying parallel to the growth direction. Various processes including competition between grains take place as growth proceeds resulting in a preferred orientation of phases within the grains with respect to the growth direction.

Because eutectics are two phase mixtures, the nature of the orientation change occuring at grain boundaries depends on the kind of preferred orientation of phases that develops in each grain. Several kinds of preferred orientation are observed in unidirectionally solidified eutectic alloys (see Table II). In each case the fibers or lamellae lie parallel to the local growth direction.

In the simplest case, only the fibers grow with a preferred crystallographic direction parallel to the growth direction and no orientation relationship exists between the fiber and matrix phases. Examples of this kind of preferred orientation are Si fibers in the Al–Si eutectic,[11] Ge fibers in the Al–Ge

TABLE II
Orientation relationships between the phases and the growth direction in unidirectionally solidified eutectics

Eutectic	Orientation relationship		Ref.
NiAl–Cr	Growth direction:	NiAl⟨010⟩, Cr⟨101⟩	
	Rods not faceted but:	NiAl(100), Cr(100)	17
Al_3Ni–Al	Growth direction:	Al_3Ni[010], Al[011] or	
	Rods not faceted at	Al[21$\bar{1}$]	21
	high growth rates but:	Al_3Ni{102}, Al{1$\bar{1}$1}	
	Horizontal growth:	G.D. Al_3Ni[010]	13
Co–Cr_7C_3	Growth direction:	α-Co[211], Cr_7C_3[0001]	
	Facet plane:	Cr_7C_3{1$\bar{1}$00}	15
Pb–Sn	Growth direction:	Pb[211], Sn[211]	
	Facet plane:	Pb(1$\bar{1}\bar{1}$), Sn(0$\bar{1}$1)	9
Al–$CuAl_2$	Growth direction:	Al[112], $CuAl_2$[215]	
	Facet plane:	Al($\bar{1}\bar{1}$1), $CuAl_2$($\bar{2}\bar{1}$1)	19

eutectic[12] and Al_3Ni fibers in the horizontally grown Al_3Ni–Al eutectic.[13] In these eutectics, fiber nucleation must be independent of the crystallographic orientation of the matrix, but rapid fiber growth is probably limited to one crystallographic direction. The alignment of fibers parallel to the growth direction may occur by competition between fibers or by fiber reorientation. Fiber reorientation can occur by successive twinning as observed in the case of the Si and Ge fibers[11,14] or by the formation of successive dislocation sub-boundaries.[13] In some eutectics with this kind of preferred orientation, the fibers may have facet planes resulting from a faceted growth mode. An example would be Al_3Ni fibers in the Al_3Ni–Al eutectic grown at low rates.[13]

Another case is where the fibers and the matrix each grow with a preferred crystallographic direction parallel to the growth direction but with no orientation relationship with respect to each other. An example of this kind of preferred orientation is the Cr_7C_3–Co eutectic.[15] In this case the matrix as well as the fibers develops a preferred orientation relative to the growth direction through competition between grains or phase reorientation.

Still another case is where the fibers or lamellae and the matrix each grow with a preferred crystallographic direction parallel to the growth direction and with an orientation relationship with respect to each other. This orientation relationship can be described by specifying a match between low index planes in each phase and a match between low index directions lying in these planes. The matching planes are normally chosen to be those corresponding to some feature of the eutectic microstructure such as the broad face of lamellae or a facet plane. The matching directions in these planes are normally chosen to be those corresponding to the growth direction. An example of this kind of preferred orientation is the Pb–Sn eutectic.[9] In this eutectic the broad face of the lamellae is parallel to $(1\bar{1}\bar{1})$ Pb and $(0\bar{1}1)$ Sn. The growth direction is parallel to [211] Pb and [211] Sn.

It has been proposed that an orientation relationship between eutectic phases arises when one phase nucleates expitaxially upon the other.[16] Observations by Jaffrey and Chadwick suggest that such relationships are established almost immediately upon initiation of growth from a chill.[13] The alignment of specific crystallographic directions in the fibers (or lamellae) and the matrix parallel to the growth direction may involve both grain competition and phase reorientation processes. Hopkins and Kraft suggest that competition establishes those grains where the broad face of the lamellae or the long axis of the fibers is aligned parallel to the growth direction.[9] In subsequent growth, a phase reorientation consisting of a continuous rotation about a direction perpendicular to the growth direction occurs in each grain. This rotation along with competition between grains establishes the preferred crystallographic orientation.

On the basis of the preceding discussion, it can be seen that if no orientation relationship exists between the constituent phases of the eutectic, then there are two types of boundaries possible. One involves a change in orientation of the matrix phase; the other, a change in orientation of the fiber phase. If either phase grows with a specific crystallographic direction parallel to the growth direction, then the possible orientation changes are limited to those that can be obtained by rotation about the growth direction. Both types of boundaries would be difficult to observe by normal metallographic examination.

If an orientation relationship exists between the fibers or lamellae and the matrix, then both phases normally change orientation in crossing a grain boundary. If the phases grow with a specific crystallographic direction parallel

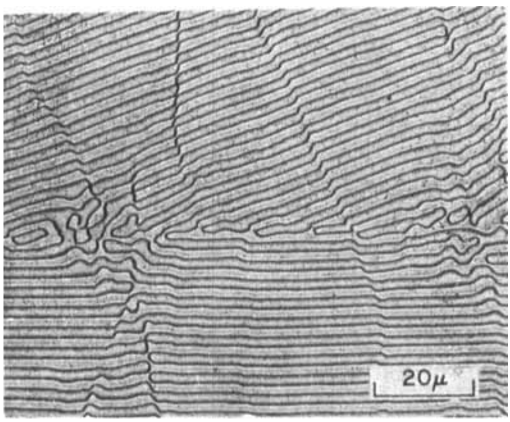

FIGURE 1 A grain boundary in unidirectionally solidified Al–$CuAl_2$ eutectic.

to the growth direction, the possible orientation changes are limited as previously described. In lamellar eutectics, grain boundaries can be readily identified by metallographic examination. A grain boundary in a unidirectionally solidified Al–Cu–Al_2 eutectic is shown in Figure 1. The preferred orientation occuring in this eutectic is described in Table II.

2.2 Interphase boundaries

The different types of interphase boundaries that are observed in unidirectionally solidified eutectics are listed in Table III. In eutectics where there exists an orientation relationship between the phases, interphase boundaries may be coherent, semi-coherent or cusp-oriented. In eutectics where no orientation relationship exists the phase boundaries are generally noncoherent.

Fully coherent interphase boundaries join phases with closely related crystal structures and matching lattice parameters. They are boundaries

TABLE III
Interphase boundary types in unidirectionally solidified eutectic alloys

Interface	Structure	Interface energy
Coherent	Lattice match at interface. Lattice planes are continuous across interface	Low
Semi-coherent	Regions of match separated by regions of mismatch (dislocations)	Intermediate
Noncoherent	No match	High
Cusp-oriented	High density of coincidence sites	Intermediate

where the arrangement of atoms in the two phases match and across which lattice planes and directions are continuous. Alloys of the NiAl–34 at. pct. Cr eutectic with additions of Mo provide an example of interphase boundaries with varying degrees of coherency.[17] The dispersed phase in these alloys is a disordered bcc solid solution of Mo in Cr while the matrix is the bcc ordered phase NiAl (CsCl structure). The alloy is fully coherent at a concentration of ~0.6 at. pct. Mo, 33.4 at. pct. Cr.

Semi-coherent boundaries join phases with closely related crystal structures, but with lattice parameters that differ significantly. Such interfaces are made up of regions of forced elastic coherency bounded by dislocations that are present to relieve the large elastic strains that would otherwise exist. The NiAl–34 at. pct. Cr eutectic provides an example of semi-coherent interphase boundaries. Observations of interface dislocations in this alloy have been reported by Walter, Cline and Koch.[18]

Cusp-oriented interphase boundaries join phases with dissimilar crystal structures. They are boundaries where the arrangement of atoms match to some degree so as to produce a large number of coincidence sites. Kraft has proposed that a criterion for predicting this type of boundary is that the matching atomic planes (or in some cases "puckered planes") in each phase have similar atom densities.[19] This criterion provides a satisfactory explanation for the interphase boundary planes observed in a number of lamellar eutectics including $CuAl_2$–Al,[19] Mg_2Sn–Mg[20] and Pb–Sn.[9] The term cusp-oriented refers to a plot of specific interface boundary energy versus the relative orientation of the phases. It is assumed that the special boundaries that possess a large number of coincidence sites would have a lower specific energy than those that do not and would correspond, therefore, to cusps in the specific energy versus orientation curve.

Noncoherent interphase boundaries also join phases with dissimilar structures. There is no continuity across these boundaries, the structure of which is completely disordered. The Co–Cr_7C_3 eutectic provides an example of this type of boundary.[15]

The specific interphase boundary energy is considered to be high for noncoherent boundaries, intermediate for semi-coherent boundaries and low for coherent boundaries. The specific energy of cusp-oriented boundaries is considered to be intermediate in magnitude.

III INFLUENCE OF INTERFACES ON PROPERTIES

3.1 Grain boundaries

There is little evidence that grain boundaries have an adverse effect on the mechanical properties of unidirectionally solidified eutectics. They normally appear to play a passive role in fracture processes acting neither as preferred initiation sites for cracks nor easy crack propagation paths. It should be pointed out that most investigators have concentrated on determining tensile properties parallel to the growth direction. As previously discussed, unidirectionally solidified eutectics have a coarse columnar grain structure aligned parallel to the growth direction so that the total grain boundary area lying perpendicular to the growth direction is small. Thus, in a tensile specimen with axis parallel to the growth direction, only small areas of grain boundary are subjected to tensile loading and they do not provide a continuous fracture path.

3.2 Interphase boundaries

Interphase boundaries affect the mechanical properties of unidirectionally solidified eutectics in two important ways. First, they are interfaces across which load is transferred betwen the matrix and the fiber. For fiber reinforcement to occur, these boundaries must be strong enough to transmit the strong shear stresses that develop in the vicinity of the fiber tips. Second, the magnitude and anisotropy of the specific interface energy largely determines the morphological stability of these alloys with respect to normal coarsening. The coarsening process results in a deterioration of mechanical properties through a reduction of constraint and dispersion hardening effects in the matrix and a decrease in the aspect ratio of the dispersed phase.

3.2.1 Load transfer

The strength of a unidirectionally solidified eutectic composite depends on the capacity of the interphase boundary region to transfer stress from the matrix to the dispersed phase. This capacity can be reduced through plastic deformation of the matrix or interphase boundary sliding. In unidirectionally solidified eutectics, the scale is such that plastic deformation of the matrix and interphase boundary sliding should not be treated as independent processes. In what follows, a dislocation model for these processes will be discussed.

To understand the plastic relaxation processes that might occur in the interphase boundary region of a eutectic composite, it is helpful to have an equation that gives the free energy change per unit volume in a uniaxially stressed body loaded uniformly by surface tractions due to changes in modulus, residual strain, dimensions and anelastic energy (i.e., changes in interphase boundary area, length of dislocation cores, etc.). The equation is

$$\Delta G' = \frac{1}{E_c^2}\left(\frac{F}{A}\right)^2 \mathrm{d}E_c + \frac{1}{V}\int (\sigma_i^o\epsilon_i^o - \sigma_i\epsilon_i)\,\mathrm{d}V + \frac{1}{V}F(L - L_o) + \Delta U_A \qquad (1)$$

where $\Delta G'$ is the free energy change per unit volume, E_c is the composite modulus, F is the total force uniformly distributed at the ends of the body, V is volume, $\{\sigma_i\epsilon_i\}$ and $\{\sigma_i^o\epsilon_i^o\}$ are the final and initial fields of residual stress and strain, respectively, L and L_o are the final and initial lengths of the body and ΔU_A is the total change in anelastic energy. The first term is the change in deformation potential and is equal in magnitude but opposite in sign to the change in strain energy due to the applied stress. It includes the change in strain energy due to the applied stress and the boundary work resulting from a composite modulus change. The second term is the change in residual strain energy. The third term is the boundary work term associated with the change in stress free length and the fourth term is the total change in anelastic energy.

Let us now consider the plastic processes that can occur in the interphase boundary region on the basis of Eq. (1).

i) Relaxation of residual stresses First we consider plastic processes that can occur in the interphase boundary region to relax residual strains in the eutectic composite. The effect of such residual stresses on the proportional limit has been investigated previously by Koss and Copley,[22] Pattnaik and Lawley[23] and Rhodes and Garmong.[24] Figure 2a shows the residual strain distribution around an isolated fiber after cooling from the eutectic temperature, while Figure 2b shows how this strain can be relieved by the formation of a prismatic dislocation loop. The energetics of this process are described by second and fourth terms of Eq. (1). It should occur if the decrease in

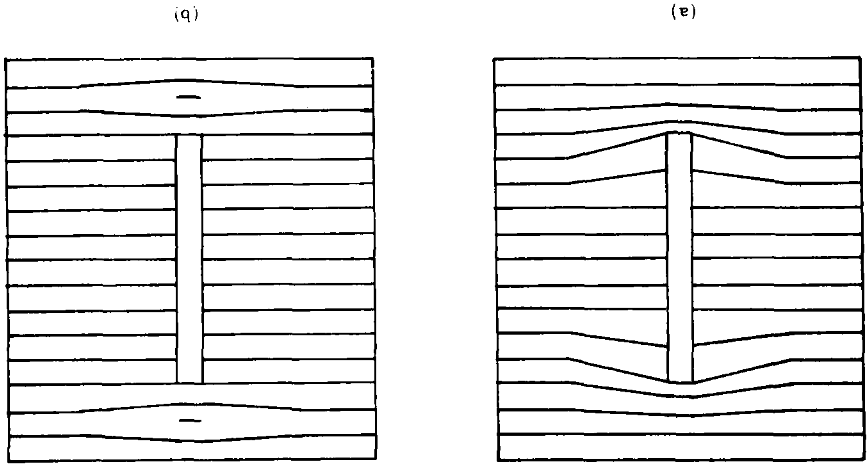

FIGURE 2 Strain distribution around an isolated fiber (a) after cooling from the eutectic temperature, (b) after the formation of a prismatic loop.

residual strain energy of the matrix and fiber is greater than the increase in strain energy and anelastic energy due to the prismatic dislocation loop. Whether or not this type of process can occur also depends on crystallographic considerations. An example where such relief appears likely is the Al_3Ni–Al eutectic. In this eutectic, the fiber axis lies parallel to the Burgers vector for slip in the fcc aluminum matrix and the facet planes of the fiber which are observed in eutectics grown at low rates lie parallel to the matrix slip planes.[21] It should be recognized that stress relief can result from the formation of any dislocation loop of the appropriate sign with a component of Burgers vector lying parallel to the fiber axis. The energetics for this process are the same as for the case of the prismatic loop.

ii) Interaction of glide dislocations with fibers A possible source of stress relaxation would appear to be the interaction of matrix glide dislocations with the interphase boundary. Figure 3 shows the example of a dislocation loop left at the interphase boundary of a fiber by a dislocation that has

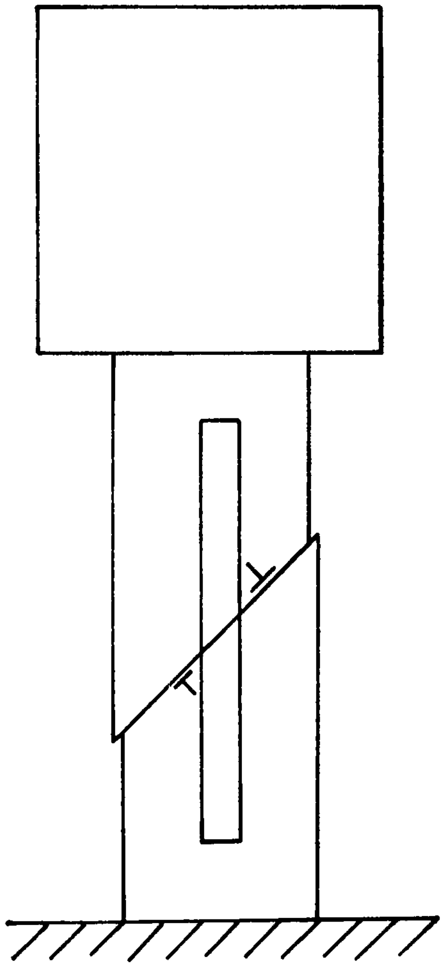

FIGURE 3 A dislocation loop left at the interphase boundary of a fiber by a dislocation that has sheared the surrounding matrix.

completely sheared the surrounding matrix. The energy change for this process is described by the second, third and fourth terms in Eq. (1). The process will occur if the energy decrease due to the boundary work resulting from the change in stress free length exceeds the increase in an elastic and residual strain energy as the dislocation bows around the fiber. The nature of the interaction between the loop and the fiber depends on the angle between the Burgers vector and the fiber axis. The component of Burgers vector lying perpendicular to the fiber axis acts to shear the fiber or to crack the interphase boundary while the component lying parallel to the fiber axis acts to increase the compressive stress in the fiber and thus the shear stress at the interphase boundary near the fiber tips. Dislocations with Burgers vector oriented approximately parallel or perpendicular to the fibers will probably not be involved in the plastic deformation of the matrix because the Schmid factor for such dislocations must be close to zero.

iii) Relaxation of interface shears due to applied load Figure 4 shows the distribution of strain surrounding an isolated fiber for several loading conditions. Figure 4a shows the region around the fiber before loading while

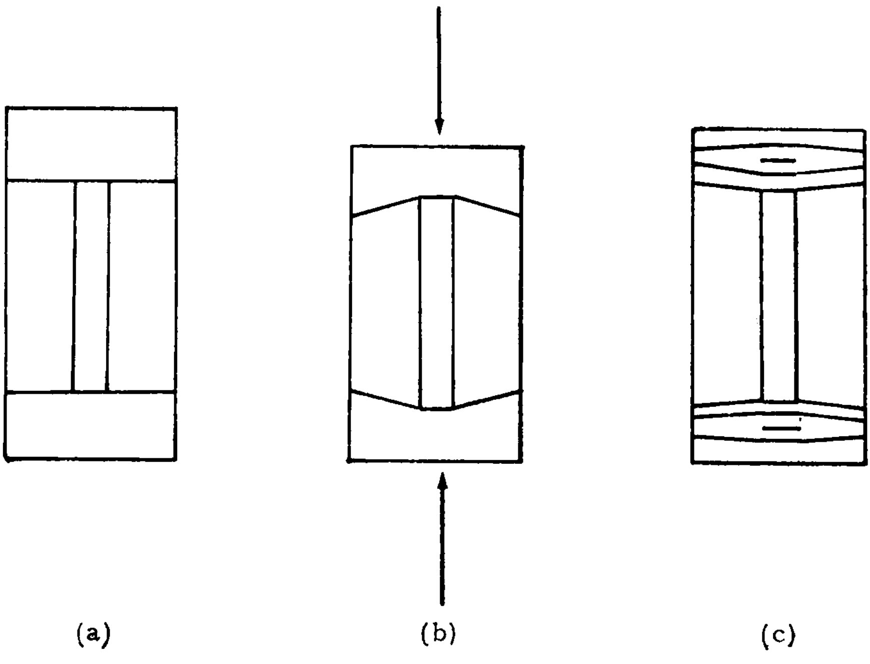

FIGURE 4 Strain distribution around an isolated fiber for several loading conditions: (a) before composite is loaded (b) after loading, and (c) after the formation of a prismatic loop (unloaded).

Figure 4b shows the same region after the load is applied. As previously discussed, shear stresses at the fiber tip can be relieved by the formation of prismatic loops, Figure 4c. In this case, the Burgers vector of the loops must be close to parallel to the fiber axis. This is because loops of the proper sign to relieve the shear stresses at the fiber tip will have to move against the resolved shear stress due to the applied stress. The energetics for this process involves the second, third and fourth terms. The process will occur if the energy decrease due to the boundary work term resulting from the change in stress free length, the change in residual strain energy due to the change in misfit between the fiber and the matrix and the formation of the dislocation loop and the change in anharmonic energy due to the dislocation core sum to give a net decrease in free energy. The presence of a residual strain due to thermal expansion mismatch prior to loading can obviously influence the likelihood of this process depending on the sense of the residual and applied stress.

In summary, some understanding of stress relaxation processes in eutectics can be obtained from a dislocation model approach. It is clear that interactions with glide dislocations act to increase the shear stress at the fiber tips. If crystallographic factors are favorable, the formation of prismatic loops may act to relieve shear stress at the fiber tips arising from thermal expansion mismatch or from the applied stress.

3.2.2 Coarsening

In eutectic coarsening, the free energy of the alloy decreases as a result of changes in total interphase boundary energy and residual strain energy. The total volume fraction of fibers (or lamellae) remains constant while there are changes in fiber size, distribution and shape. Smartt, Tu and Courtney have investigated the kinetics of coarsening of the Al–Al_3Ni eutectic.[25] They describe the aligned fiber structure as intrinsically morphologically stable under elevated temperature exposure. They proposed that fiber coarsening occurs in three stages. The first stage is described as a two-dimensional Ostwald ripening process. During this stage the cross-sectional shape of the fiber is constant while the average fiber diameter increases, as theoretically predicted,[26] according to a one-third power time law. This is followed by a stage during which the fiber diameter remains nearly constant. There is a final stage where coarsening resumes after a breakdown of the stable structure by a fault propagation process similar to that suggested by Graham and Kraft.[27] In this final stage of coarsening extensive fiber shortening takes place and the fibers are observed to develop facets. A study of the crystallography of faceting in Al–Al_3Ni has been made by Garmong, Rhodes and Spurling.[21]

In the case of Al–Al_3–Ni, strain energy does not appear to have much influence on the coarsening process. Throughout the process the main change in free energy would appear to result from the decrease in total interphase boundary energy. This may be because at temperatures sufficient for coarsening to occur the residual stresses have been relaxed.

From the preceding discussion, it can be seen that the magnitude of the specific surface energy should have an important influence on the kinetics of coarsening. All other factors equal, coarsening rates should be greatest in eutectics with noncoherent interphase boundaries and least in those with coherent boundaries. Semi-coherent and cusp-oriented boundaries represent intermediate cases. Additional stability may occur in cusp-oriented interphase boundaries because of the anisotropy of the specific energy. This anisotropy should give additional stability to faceted surfaces with respect to shape perturbations.

Finally, it should be recognized that eutectics with cusp-oriented interfaces may exhibit a special type of instability when unidirectionally solidified to form an irregular shape. This is because the fibers (or lamellae) when growing through an orafice or under conditions where the growth direction must change may not be able to grow with their normal cusp-oriented interfaces. According to Hunt and Chilton, this can result in regions of irregular structure where the dispersed phase is bounded by noncoherent boundaries.[28] These regions would have a lower resistance to coarsening than the bulk of the alloy and thus after exposure at high temperatures would be likely regions for the initiation of failure.

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