Boundary Stability Under Nonequilibrium Conditions.

Final Report

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Morphological (Diffusional) Stability.

1. Stability analysis of the spheroidization of rod-like precipitates, pores or reinforcement

Previous investigations of the Rayleigh spheroidization process have predicted a shapeinstability mechanism for a cylindrical rod where a rod splits into a series of spheres if the surface wavelength is greater than the initial rod circumference. A critical point in previous studies is that there exists a surface wavelength at which the growth rate reaches the maximum and that this would be the experimentally observed wavelength of the instability. There is a growing body of experimental data from ceramic and metals processing studies which reveals a considerable scatter in the growing wavelengths from the one predicted by “the maximum growth hypothesis.”

Our study was carried out using in situ TEM to study shape evolution on free standing rods. The analysis of the experimental results using a linear multi-harmonic initial surface waves with analytical methods, as compared to a single harmonic in the classical approach, and using numerical techniques to examine nonlinear effects. The major result has been the discovery that the observed wavelength of spheroidization depends on the stochastic nature of the initial perturbations on the rod surface.

2. Shape evolution in a bamboo structure

The finite difference and an analytical Fourier study on the shape evolution in an initially-cylindrical bamboo structure (a cylindrical rod cut by grain boundaries) was carried out under the usually assumed conditions of constant volume and constant length. The discrepancy in these studies suggest that the constraint of constant length should be removed, and new boundary conditions of puckering or lateral movement of grain boundaries would be more realistic for modelling shape evolution in a bamboo structure.


A. Research accomplished.

In the past year, we have developed a novel technique, termed the Discrete Atom Method (DAM), which is predicated upon Hookean atomic interaction and Monte Carlo diffusion. Because of the simplicity, minimal hardware requirements, speed and apparent accuracy of the new approach, this is a major breakthrough in the computational modelling of materials interface behavior. Over the past year, the powerful applicability of the DAM was demonstrated by the modelling of several difficult and unsolved elasticity problems. The problems treated were (1) the elastic strain energy of a precipitate, (2) elastic interaction energy between precipitates, (3) shape evolution of a precipitate, (4) surface instability of an epitaxiaily-strained thin film, (5) coherency-influenced coarsening, (6) effects of externally applied stresses, and (7) interaction between a precipitate and an interphase boundary.

The DAM method is the first technique which has been able to solve the microstructural development problem of inhomogeneous elastically stressed two-phase systems. Such problems are of great practical interest and fundamentally different from that of unstressed systems. Two well-
known cases are the experimentally observed development of strong spatial correlations during coarsening of misfitting coherent precipitates in nickel-base alloys, and the observed formation of islands during a thin film processing. To understand such problems, micromechanists have long searched for a simple computational technique, through which one can analyze the elastic state associated with arbitrarily-shaped, coherent precipitates whose elastic constants are different from those of the matrix phase. Eshelby was the pioneer in the field of coherency strain, who devised the classic equivalency method; however, the method had to be limited to one ellipsoidal precipitate. Since his work, several numerical techniques have been developed, but most involve either computations of an elastically homogeneous state, i.e., with the assumption of uniform elastic constants, or approximate solutions for integro-differential equations when faced with an inhomogeneous system. The DAM method is a significant improvement on these approaches.

A DAM result is shown in Fig. 1, where the precipitate has 5% misfit strain and its shear modulus is equal to one half of the matrix phase in a plane strain condition. The numbers indicate the time in units of a million Monte Carlo diffusion attempts. An interesting feature is that shape transition begins with interfacial waves induced by the coherency strain. The waves then develop small lobes, which coarsen into a lower density of larger lobes. Some lobes eventually disappear as the equilibrium shape, balanced between the elastic strain and interfacial surface energy, is approached. Isotropic elasticity is considered in Fig. 1. Anisotropic effects are found to suppress some of the interfacial waves. In Fig. 2, the anisotropic ratio is taken to be a value similar to that of nickel, and the elastic constants of the matrix phase are twice those of the precipitate. The soft <100> directions are marked with arrows. Before achieving its equilibrium shape stretched along the soft [010] direction, the precipitate is shown to undergo a series of symmetry-breaking shape transitions from a radial to a four-fold, then to a two-fold symmetry. The evolution is consistent with experimental observations in a typical Ni-base superalloy. There has been a perplexing question why the morphological evolution in such a superalloy exhibits protrusions along elastically hard <110> directions. The DAM analysis has now resolved that they are a manifestation of strain-induced interfacial waves. Other DAM findings are discussed in the publications listed below.
Dynamic interactions between interfaces and dislocations play crucial roles in determining the properties of alloys or thin films, and yet our understanding of this subject has been severely limited. A major barrier impeding the progress has been the absence of a computational technique through which one can accurately analyze a general coherency state. With the DAM, we are now able to investigate the dynamic interaction between coherent precipitates and dislocations. The outcome of this research should expand our understanding of the following processes: how a precipitate changes its shape in the strain field of a dislocation, how a moving dislocation shears or deforms a coherent precipitate, and how the coherency strain of a precipitate relaxes through dislocation punching or interaction with lattice dislocations. In analyzing the dynamic interactions, we will start with edge dislocations based on a truncated Hookean interatomic potential. Since the repulsive force of such a Hookean potential is quite soft, the dislocation core size is relatively small, and yet it should be advantageous in an exploratory work as the model facilitates comparisons with continuum linear elasticity theory. Obviously, the effect of anharmonicity must be studied later by employing "realistic" interatomic potentials such as Embedded Atom potentials. Due to the complexity of the problem, at this phase of research only problems in a 2-dimension crystal will be examined in depth. Finally, with proven satisfactory results in the 2-dimensional case, the DAM algorithm will be extended to 3-dimensional coherency strain problems and interfacial stability problems as considered below.

II. A Unified Treatment of Single and Microcrystalline Film Edge Instabilities.

A. Research Accomplished.
We have been able to combine experiment and theory to generate a unified treatment of film edge instabilities in two systems which show a huge difference in observed behavior. The formation of a periodic or quasi-periodic morphology along the edges of unstable, two dimensional solid state structures (films, cracks, etc.) has been described by numerous investigators. Cylindrical rods with the axis perpendicular to the edge of the film are formed (fig. 3) as diffusion transfers mass from the region of high curvature at the edge to the relatively thick rods. This highly efficient mechanism of surface area reduction is responsible for the loss of structural integrity in a variety of films and coatings.

The cylindrical rods show a characteristic spacing for a given film thickness. However; there is an order of magnitude difference between the spacing/thickness relationship of single crystal films and of microcrystalline films. This disparity has been accounted for by the use of concepts previously developed within this research program. Using the same approach for both types of films, but accounting for the specific geometry of individual grains in the microcrystalline film predicts separate curves corresponding to the respective nature of the film. The comparison of theory and experiment is shown in figure 3 and an excellent correlation is found for both the microcrystalline data and the single crystal data.
III. Validation of the Structure Based Grain Boundary Diffusion/Migration Model.

A. Research Accomplished.
The existence of a grain boundary dislocation mechanism to link grain boundary diffusion and grain boundary migration has been debated for almost 25 years. This portion of the research program has concentrated on understanding the atomic processes involved in the migration of grain boundaries in stressed thin films as a means to test the structure based model. From preliminary experiments (M. S. Thesis, T. M. Lillo) two interesting observations were made in examining the migration of such boundaries. First, it was found that grain boundary dislocations migrated within the grain boundary plane as the boundary itself migrated. This dislocation migration was found to be nonconservative in nature, involving both glide and climb. Second, a change in thickness in the crystal behind the advancing grain boundary was also noted (fig. 4).
Cross section of film under compressive stress. Thickening at elevated temperature occurs behind the migrating grain boundary.

Fig. 4

The sign of the thickness change depended upon the nature of the applied stress; that is, thickening was observed under compression and thinning under tension. A simple model was developed in an effort to describe these observations. It was assumed, based on nonconservative dislocation motion, that there existed a flux of atoms along the grain boundary, in particular along the grain boundary dislocations, to or from the surface thus leading to the observed thickness changes. The simplified model predicted the observed thickening to be dependent upon both the initial thickness of the grains comprising the grain boundary and the grain boundary structure, i.e. number and type of grain boundary dislocations and their migration rates.

These initial observations provided the basis for the experiments and analyses leading to the Ph.D. dissertation of T. M. Lillo. Two unique aspects of Dr. Lillo's work included 1) measurement of thickness both ahead and behind the migrating grain boundary and 2) full characterization of the grain boundary structure and crystallography. Furthermore, the simplified model initially developed was treated as a moving boundary problem and incorporated boundary structure as well as initial thin film thickness. Representative experimental results from two grain boundaries examined are shown in Figs. 5 and 6. It should be noted first that the change in thickness for each of these boundaries increases as the thin film thickness increases. The thickness increase appears to be nearly linear over a fairly wide range of thin film thicknesses. The model and experimental results are further compared using the grain boundary structure. Of all possible variables involved in grain boundary migration it was determined that model predictions were most sensitive to the grain boundary structure, which describes the secondary grain boundary dislocation content. Careful crystallographic analyses coupled with O-lattice theory enabled the grain boundary structure to be determined for two of the grain boundaries studied. Comparison of the model and experiment for these boundaries is also shown in Figs. 5 and 6. It can be seen that fairly good agreement exists between theory and experiment, especially when considering the slopes of the
thickness change versus film thickness curves. This does suggest that the proposed atomic processes involved in grain boundary migration are indeed active.

Figure 5. Comparison between the experimentally determined dependence of \( \Delta t \) on the initial thickness for Boundary #5 and that predicted by the model, using the theoretical boundary structure parameter, \( Q_{\text{theory}} \), calculated from the crystallographic data for Boundary #5.

Figure 6.

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**STUDENT SUPPORT**

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Proceedings of Interface Dynamics and Growth, in press.


29. E. D. McCarty and S. A. Hackney, “Dislocation Strain Field Probes of Metal/Ceramic Interfaces”, Accepted to MRS Proceedings.


