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# ANNUAL REPORT

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# THE EFFECT OF ELASTIC STRESS ON OSTWALD RIPENING PHENOMENA

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## I. PROGRESS DURING SECOND FUNDING PERIOD

The purpose of our research program is to understand the influence of elastic misfit strain and applied stress on the development of microstructure in two-phase coherent alloys. The long-term goal of our research is to identify those material parameters that promote microstructural stability in multiphase systems at elevated temperatures.

Our efforts since the last progress report in February 1991 have been twofold. First, we have continued performing coarsening simulations of elastically and diffusionally interacting particles in an infinite matrix. In addition, we have been performing simulations of unstressed systems during coarsening. Secondly, we have begun to develop an experimental component to the research that would allow a careful comparison between theory, computer simulations and experiment.

We have continued to focus our computer simulations of stressed systems on elastically homogeneous and isotropic systems containing tetragonally misfitting particles. These assumptions allow a tremendous simplification of the elasticity problem and make the simulations tractable. They also allow the effect of misfit strain on coarsening to be discerned.

We have shown previously how elastically-induced particle migrations and preferential coarsening can influence microstructural development in systems containing misfitting particles by examining the coarsening behavior of two particles and small groups of five particles. We have since concentrated on simulating the coarsening behavior of larger groups of particles as well as systems with an "infinite" number of particles using periodic boundary conditions. Groups containing about 40 particles allow for complex elastic and diffusional interaction between particles but still allow the individual particles to be displayed in time.

Figures 1 - 4 depict the coarsening behavior of such a group of particles. Figure 1 shows the initial microstructure used for the following simulations. It is displayed as two projections, onto the x-y and x-z planes, respectively. Figure 2 shows the microstructure at the end of a simulation that coarsened only under the influence of capillarity. The particles have grown in size, decreased in number, and have not developed any specific spatial alignments.

Figure 3 depicts the final microstructure of a coarsening system containing tetragonally misfitting particles. The axis of tetragonality is along the z axis and the misfit in the z direction is slightly larger than

in the x and y directions. Although the initial microstructure is identical to that used to obtain Fig. 2, the final microstructures are much different. There is a highly pronounced alignment of the surviving particles. Of the five surviving particles, four are aligned in (or near) a plane perpendicular to the z axis. This configuration of particles gives a lower elastic energy since particles aligned perpendicular to the z axis have a negative interaction energy in this case.

Figure 4 also depicts the final microstructure obtained by simulating the coarsening behavior of tetragonally misfitting particles. However, in this case, the misfit in the z direction is slightly less than that in the x and y directions. The particles align themselves along the z axis, instead of perpendicular to it as in Fig. 3, reflecting the negative elastic interaction between particles in this orientation. It is also interesting to note that the remaining particles in each of the simulations depicted by Figs. 2 - 4 are different.

The technique of irreducible multipole tensors employed in solving the coupled elasticity and diffusion problem is also applicable to unstressed systems. By considering higher-order multipoles, the technique allows a more accurate simulation of coarsening behavior than the heretofore used monopole solution. In addition, it allows for particle migration and permits the development of small spatial correlations between particles to be followed, if they do indeed occur in unstressed systems. Therefore, we have been using this technique to simulate coarsening behavior in unstressed systems at small particle volume fractions (0 - 4%) in order to compare our simulations with previous coarsening simulations and as a check on the numerical work.

To date, our simulations show that these small, non-zero volume fraction systems exhibit scaling properties and that the average particle size increases proportional to $t^{1/3}$. The time-independent particle size distribution broadens as the volume fraction increases, in agreement with other simulations. Particle correlations are also found to develop with time, corroborating the predictions of Marder's theory. Small particles tend to be near large particles and vice versa. Our coarsening rate constants are higher than those predicted by other simulations, except for the work of Marder. We are unable to explain these differences at this time.

We are in the preliminary stages of designing experiments to investigate how the sign and magnitude of the misfit strain and the presence of an applied stress influence microstructural evolution during coarsening. The development of spatial correlations and the effect of stress on the coarsening kinetics will

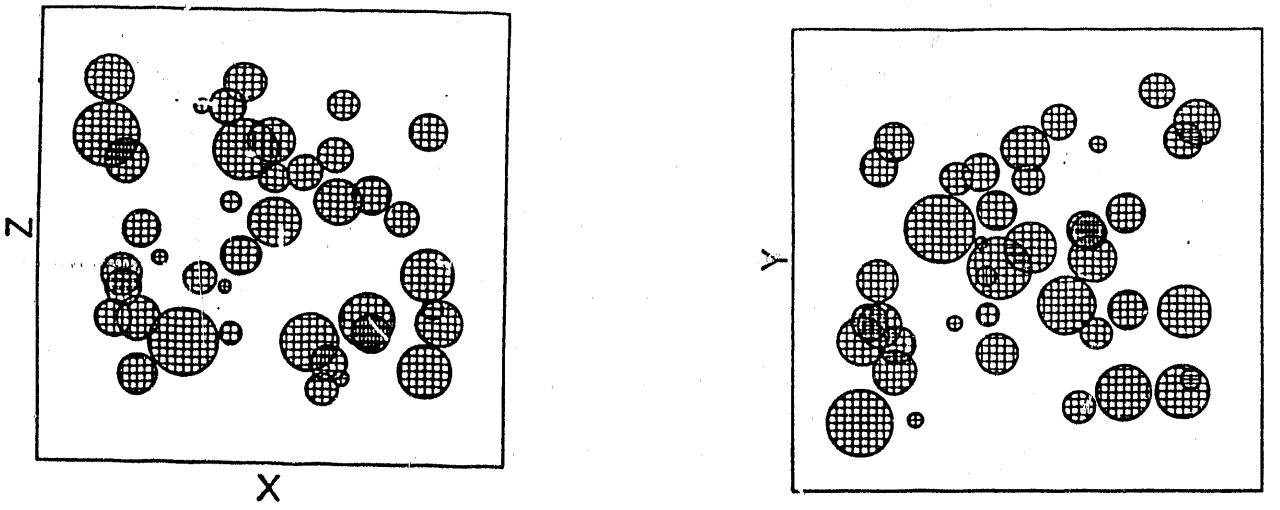


Fig. 1 - Initial configuration projected onto the x-y and x-z planes.

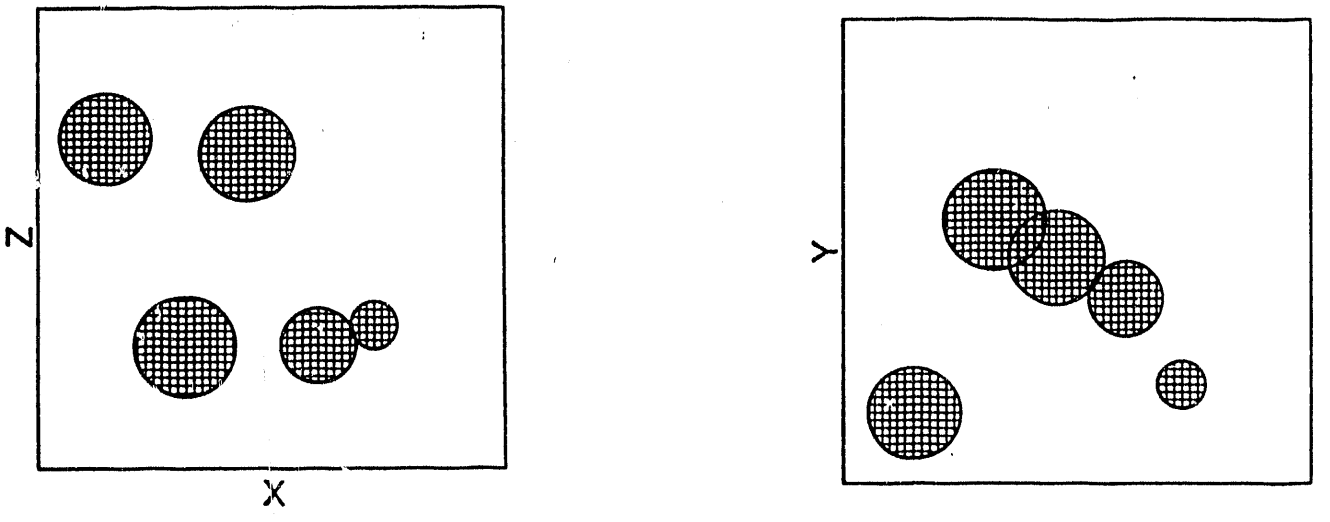


Fig. 2 - Microstructure after coarsening simulations in the absence of all stress effects. (capillarity-driven coarsening)

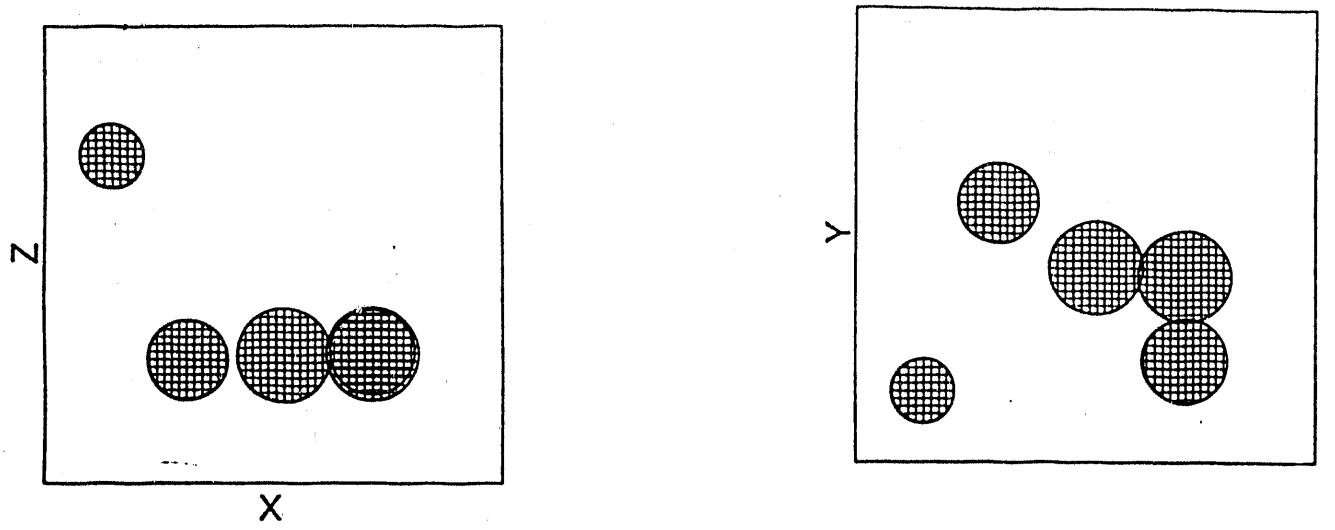


Fig. 3 - Final microstructure after coarsening simulations for tetragonally misfitting particles. Misfit in Z direction is slightly greater than in the x and y directions.

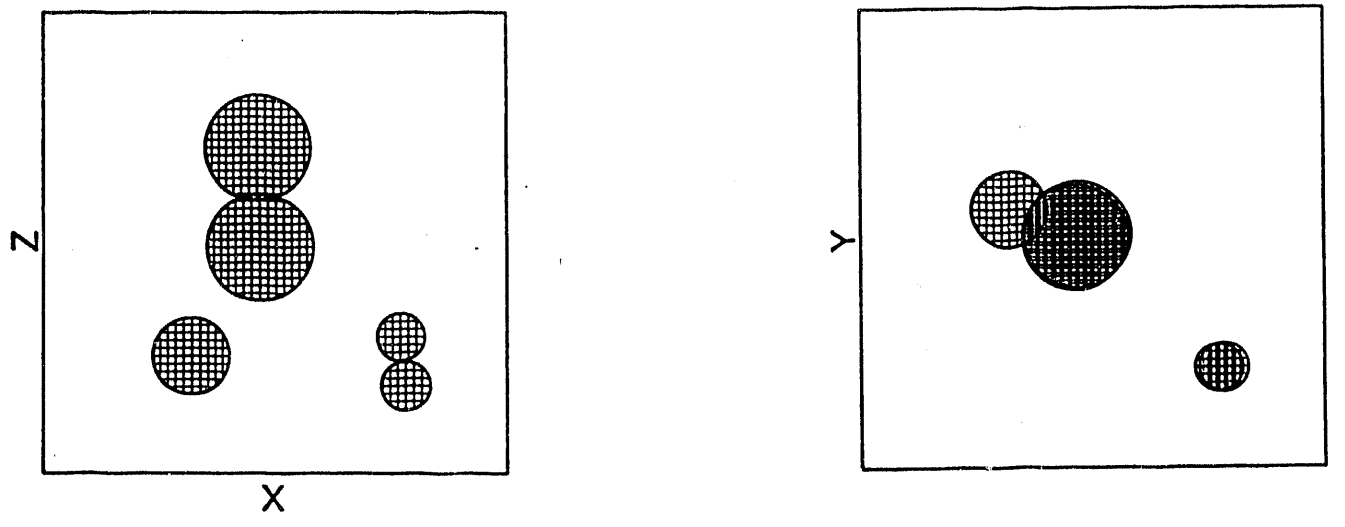


Fig. 4 - Final microstructure after coarsening simulations for tetragonally misfitting particles. Misfit in z direction is slightly less than in x and y directions.

be examined.

It is intended for the experiments to be compared with the simulations. We have chosen Ni-Al with ternary additions of Mo as a model alloy. The Mo segregates preferentially to the matrix phase and is used to control the particle misfit. We are currently developing alloys with a $\gamma$ volume fraction of approximately 10% with particle misfits ranging from about 0.5% to -0.5% at room temperature by changing the Mo concentration. The three initial alloy compositions being examined are, in atomic percent, $Ni_{85}Al_{10}Mo_5$, $Ni_{84}Al_6Mo_{10}$ and $Ni_{81}Al_5Mo_{14}$. Misfit strains and volume fractions at temperature need to be verified before single crystals of the alloys are obtained.

## II. PROPOSED WORK FOR THE NEXT FUNDING YEAR

During the coming year, we will continue our simulations of stressed and unstressed systems. One important goal is to independently verify the results of our simulations of systems containing large numbers of particles per basis with periodic boundary conditions. We will first concentrate on unstressed systems. The diffusional cut-off distances will be extended. If necessary, we will develop new code as a further check on existing code. The verification is important both as a check on the existing code for the stressed case and for illuminating the coarsening behavior of unstressed systems. Our simulations are the only ones to date that allow for particle migrations and higher-order multipoles. Since our simulations of unstressed systems are, thus far, the only simulations to support the theoretical work of Marder, we may have shown that one must allow for both the development of spatial correlations and particle migrations if the coarsening behavior of unstressed systems (including the prediction of rate constants) is to be adequately predicted. This work will be undertaken by W.Hort, a new graduate student, with interaction from former graduate student Dr. T. A. Abinandanan.

We will establish a procedure for examining the coarsening behavior of systems subjected to an external elastic stress. A major difficulty in performing such simulations is obtaining a reasonable approximation to the elastic field. Such an approximation must be accurate enough to allow the diffusion problem to be solved yet simple enough to implement at each time step of the simulation. Since the difference in elastic constants between particles and matrix is important in determining the development of microstructure in systems subject to external stress, we cannot use a superposition of particle elastic fields in a homogeneous system as in the present simulations. Instead, we will try to obtain an approximation to the elastic fields using an integral equation. We would like to simulate the coarsening behavior of such systems for comparison with some of the planned experiments.

As indicated, major emphasis is being placed on the design and implementation of experiments to examine coarsening behavior in stressed systems. Once we have determined the appropriate alloy compositions, we will order single crystals for use in both externally stressed and unstressed systems. Dr. Michael Fahrmann, a visiting scientist from Germany, has undertaken these preliminary experiments. In collaboration with Dr. Peter Fratzl at the University of Vienna, we intend to use small angle scattering techniques to obtain average particle sizes as a function of time. In addition, we hope to quantify the spatial correlations at the earlier stages of coarsening. TEM work will also be performed on all specimens, in collaboration with Prof. Pollock.

We hope to begin exploratory experiments of coarsening in oxides with Profs. M. Ready and T. Pollock of CMU. Model systems include the binary partially stabilized zirconias. The particles possess a tetragonal misfit, which might allow a more direct comparison to the simulations currently being performed. In addition, the size and distribution of the second-phase particles is critical for the mechanical behavior of the system.

## III. PERSONNEL

From May 1, 1990 to March 31, 1991, I was a visiting Professor in the Institut fur Metallforschung at the Technical University in Berlin. During this time I interacted with a group headed by Prof. Feller-Kniepmeier which has been examining microstructural evolution in the nickel-based super alloys. From April 1, 1991 to June 30, 1991, I was an Alexander von Humboldt Fellow and a guest of Prof. H. Schmalzried at the University of Hannover where we began research on the influence of elastic stress on solid state reactions in ceramic systems. This joint research is continuing. I returned full time to Carnegie Mellon on July 1, 1991. I was also the recipient of the 1990 Outstanding Paper Award in Acta Metallurgica et Materialia.

T.A. Abinandanan, a graduate student supported by this grant, earned his Ph.D. degree in April 1991. A copy of his thesis is enclosed. He did an outstanding job and is currently doing post-doctoral work with Dr. Georges Martin (CENS) in Saclay, France.

Werner Hort is a new graduate student as of September 1991. He has the equivalent of an M.S. degree in physics from the University of Saarland and has performed computer simulations. He makes a strong addition to the group.

Dr. Michael Fahrmann is a visiting scientist from Germany and joined us in February 1992. He receives half of his support from the Alexander von Humboldt Foundation and half from this grant. He has a strong experimental background and is actively involved in our experiments.

In addition, we are beginning to collaborate with Dr. Peter Fratzl of the University of Vienna and Profs. Tresa Pollock and Michael Ready of Carnegie Mellon. Dr. Fratzl will be involved in the small angle x-ray analysis. He is well known for his work on small angle scattering and analysis of scaling effects during coarsening. Prof. Pollock has been actively investigating the high-temperature deformation and microstructural evolution of the nickel-base superalloys. Prof. Ready is a ceramist interested in the mechanical behavior and phase transformations of zirconia. Profs. Pollock, Ready and I are attempting to extend some of the techniques developed in this work to ceramic systems. They receive no compensation from this grant.

## IV. PUBLICATIONS

The following publications resulting from research supported by this grant have been published or submitted since July 1991:

1. P.W. Voorhees, G.B. McFadden, and W.C. Johnson, "On the Morphological Development of Second-Phase Particles in Elastically Stressed Solids", accepted Acta Metall. et Mater.
2. T.A. Abinandanan and W. C. Johnson, "Coarsening of Elastically Interacting Coherent Particles: 1. Theoretical Formulation", submitted.
3. T.A. Abinandanan and W. C. Johnson, "Coarsening of Elastically Interacting Coherent Particles: 2. Simulations of Preferential Coarsening and Particle Migrations", submitted.
4. W.C. Johnson, T.A. Abinandanan and P.W. Voorhees, "Influence of Elastic Stress on Microstructural Evolution during Ostwald Ripening", Proceedings International Workshop on Computational Materials Science, Japan NRIM, 95-102, 1991.
5. W.C. Johnson and P.W. Voorhees, "Elastically Induced Precipitate Shape Transitions in Solids", in Nonlinear Phenomena, vol.2, eds. G. Martin and L. Kubin, 1992.
6. S. Satoh and W.C. Johnson, "Elastically Induced Precipitate Shape Transitions in a Cu-2at% Co Alloy", accepted with mandatory revisions, Metall. Trans.

## V. SEMINARS

Since the last progress update in February 1991, W.C. Johnson has presented the following seminars related directly to this grant:

1. "Influence of Misfit Strain on Coarsening", A.V. Luikov Institute for Heat and Mass Transfer, BSSR Academy of Sciences, Minsk, USSR, March 6, 1991. (Invited)
2. "Phase Equilibria in Stressed Systems", Institute for Physical Chemistry and Electrochemistry, University of Hannover, Germany, June 11, 1991. (Invited)
3. "Coarsening in Elastically Stressed Systems", German Physical Society Meeting, Munster,

Germany, April 8,1991.

4. "Role of Elastic Interactions During Coarsening of Coherent Particles", TMS-AIME Fall Meeting, Cincinnati, Ohio, October 24, 1991.

He has been asked to present the following invited seminars:

1. "Influence of Misfit Strain on Ostwald Ripening", Depa tment of Materials Science, University of Virginia, Charlottesville, VA, April 13, 1992. (Invited)
2. "Influence of Misfit Strain on Ostwald Ripening", International Centre for Mathematical Sciences, Workshop on Kinetics of Phase Transformations, Edinburgh, Scotland, August 5-10, 1992. (Invited)
3. "Ostwald Ripening in Elastically Stressed Systems", Workshop on Mathematics in Materials Sciences, Virginia, September 21-23, 1992. (Invited)
4. "Ostwald Ripening in Elastically Stressed Systems", TMS-AIME Fall Meeting, Chicago, IL, November, 1992. (Invited)

T.A. Abinandanan has presented the following invited seminars related directly to this grant:

1. "Role of Elastic Interactions During Coarsening of Coherent Precipitates", CENS, Saclay, France, July, 1991.
2. "Role of Elastic Interactions During Coarsening of Coherent Precipitates", Institute for Physical Metallurgy, Gottingen, Germany, April, 1992.

## VI. CURRENT AND PENDING SUPPORT

| | |
|---|---|
| Title: | Thermodynamics of Nonhydrostatically Stressed Solids and Interfaces |
| Sponsor: | National Science Foundation |
| Duration: | 1 August 1990 to 31 July 1992 |
| Status: | current - creativity extension |
| Amount: | $189,043 |
| Effort: | 1 Su, 1/2 AY |

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|---|---|
| Title: | Thermodynamics of Crystalline Solids |
| Sponsor: | National Science Foundation |
| Duration: | 1 August 1992 to 31 July 1995 |
| Status: | pending |
| Amount: | $304,427 |
| Effort: | 1 Su, 1 AY |

# END

# DATE FILMED

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