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AN UNSOLICITED PROPOSAL FOR A COOPERATIVE AGREEMENT
BETWEEN NASA JOHN GLENN RESEARCH CENTER AND
THE UNIVERSITY OF AKRON
AKRON, OHIO 44325-2102

PROJECT TITLE:
THEORY, MODELING, SOFTWARE AND
HARDWARE DEVELOPMENT FOR ANALYTICAL
AND COMPUTATIONAL MATERIALS SCIENCE

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1 Introduction

The focus of this Cooperative Agreement between the Computational Materials Laboratory (CML) of the Processing Science and Technology Branch of the NASA Glenn Research Center (GRC) and the Department of Theoretical and Applied Mathematics at The University of Akron was in the areas of system development of the CML workstation environment, modeling of microgravity and earth-based material processing systems, and joint activities in laboratory projects. These efforts complement each other as the majority of the modeling work involves numerical computations to support laboratory investigations.

Coordination and interaction between the modelers, system analysts, and laboratory personnel are essential toward providing the most effective simulations and communication of the simulation results. Toward these means, The University of Akron personnel involved in the agreement worked at the Applied Mathematics Research Laboratory (AMRL) in the Department of Theoretical and Applied Mathematics while maintaining a close relationship with the personnel of the Computational Materials Laboratory at GRC. Network communication between both sites has been established. Below is a summary of the projects we undertook during the time period 9/1/03 - 6/30/04.

2 System Development at NASA GRC

The primary goal was continued integration of the Computational Materials Laboratory into a smoothly working unit capable of timely response to different needs. Hence, the system management team investigated, recommended, and maintained state-of-the-art equipment such as workstations, printers, disk drives, memory, graphics cards, monitors, CPU upgrades, keyboards, ethernet cards, and tape back-up systems. The team managed network services including printing, user accounting, server management, hardware and software installation, backups, and data recovery. Software packages, such as FLUENT, FIDAP, POLYFLOW, and CFD-ACE were maintained. Additionally, the team investigated and evaluated new software and hardware as these became available, making recommendations as to which should be acquired to enhance the capabilities of the CML and the AMRL at The University of Akron.

3 System Development at The University of Akron

For the co-operative agreement to be fruitful we maintained a similar setup as at the CML. We maintained constant network connectivity within a heterogeneous network of systems, using Win-
dows 2000, UNIX, and Linux operating systems. We managed network services including printing, user accounting, server management, hardware and software installation, backups, and data recovery. Finally, the system management dealt with all issues relating to security, both external and internal, between nodes on the network.

In the area of technical support maintained the role as primary technical support for both professors and students involved in this cooperative agreement. This role included fixing any software problems that users encounter and teaching new and existing users how to use the systems.

4 Modeling at NASA GRC

The research effort under this joint cooperative agreement centered on numerical simulations of both microgravity and terrestrial processes and material processing systems. Specific projects in this effort were determined in consultation with our grant technical officer, Dr. Arnon Chait.

5 Modeling at The University of Akron

Solution Methodologies – The modeling team at The University of Akron uses a combination of modern mathematical techniques coupled with practical computational methods to simulate processing systems. Primarily we employ asymptotic and perturbation techniques to develop analytical solutions to the systems of equations governing these processing techniques. This approach enables one to represent the general trends that material and system parameters have on the system performance. The solutions derived by these procedures are quantitatively accurate in a range of application and may give insight into the system behavior outside of this range. Hence, this approach complements full numerical simulations being performed at NASA GRC with the combined approach of the two methodologies yielding the most complete analysis. This approach also allows one to investigate a variety of models in response to changing technologies, and can be modified to new areas of application.

Objectives – While members of the modeling team did not receive any direct monetary support from this cooperative agreement, the members did make use of the AMRL facilities that were partially supported by this agreement. Hence, we describe the modeling projects undertaken during the time period of the cooperative agreement.

The following projects concern the development and application of mathematical methods for the formulation, analysis and solution of material processing system models. Specifically, we examined time-dependent sharp-interface solidification models, phase-field models, nanoscale models of
scanned probe oxidation, and models for the coating and applications of nanofibers. The general goals of the projects are to examine simple model problems to determine the process sensitivity to the operating conditions, describe the scientific phenomena of heat, mass, and momentum transport within the system, and to elucidate the coupled dynamics of these transport mechanisms. During the time frame of this effort we conducted work for:

- an asymptotic approach for investigation of the time varying evolution of solidifying fronts and concentration profiles in a two-dimensional configuration including side wall heat transfer, anisotropy, and attachment kinetics,

- a multi-parameter asymptotic expansion approach for investigation of phase-field models. By expanding in both the interface thickness and another relevant parameter, such as aspect ratio, we find the leading order problem will recover the sharp-interface model developed above and the next correction will include the effects of non-zero interface thickness.

- preliminary development of an investigation that couples mathematical modeling and analysis to study lithographic patterning techniques based on scanned probe oxidation at the nanoscale.

- preliminary development of models for the investigation of the coating of polymer nanofibers with conducting materials, and the subsequent response of the coated nanofibers (or newly formed nanotubes after dissolution of the nanofiber core) to applied electromagnetic fields.

*Time-Dependent, Sharp-Interface, Directional Solidification Models* – There has been an extensive amount of investigation of Stefan-type problems and directional solidification configurations in the literature. Exact solutions to these classes of problems are generally restricted to unbounded domains and subject to limitations on the boundary conditions (for example, constant temperature). For this reason a variety of approximate analytical and numerical approaches have been developed to examine domains and boundary conditions which more closely simulate processing conditions.

Analytical approaches have generally been concerned with one-dimensional, planar interface, time-dependent models or two-dimensional, steady-state models. There have also been quasi-steady models which employ perturbation expansions in powers of a small parameter, which is inversely proportional to the latent heat, and small in magnitude due to the relatively large value of the latent heat. Almost exclusively these one-dimensional analyses have investigated pure systems over alloy systems.

The two-dimensional analytical models incorporate side-wall heat transfer and lead to more realistic non-planar solidification fronts. It is generally believed that one wishes to set the processing conditions to minimize front curvature so as to minimize transverse temperature gradients (hence,
thermal stresses) and solute segregation. These two-dimensional models give some insight into the interplay between transverse and axial heat transfer in establishing the front shape. Our earliest approaches to this problem consist of expansions in a small aspect ratio and a boundary layer analysis near the solidification front. This approach has been successful for pure and alloy systems, because most processing configurations seem to be characterized by a small aspect ratio. This approach is readily adaptable to ampoule and containerless geometries and we have used it to model a variety of processing techniques.

We combined the quasi-steady perturbation approach with the small aspect ratio approach. This enabled us to examine time-dependent, two-dimensional situations. To date we have developed a model that simulates two-dimensional, time-dependent directional solidification of a binary alloy. In this model evolution equations are developed for the leading order planar location of the front as well as for non-planar corrections to the shape of the front. The leading order solute profile is similar to that predicted by the Scheil equation. We further developed the model for an axisymmetric configuration. Undercooling, either by thermal means or by solute rejection, leads to non-planar fronts. This may be related to a transition to cellular or dendritic morphologies. This aspect will also be relevant to the phase-field projects described in the next section, since this project will potentially provide some analytic results for non-planar front solidification. We also developed a small aspect ratio, two-dimensional, time-dependent model that includes the concept of a stagnant film. The asymptotic structure of the model consists of keeping the stagnant film within the boundary layer region that develops due to the small aspect ratio. By varying the thickness of the film from zero (a well-mixed Scheil model) to beyond the boundary layer (pure diffusion model), we analytically investigated the various regimes of solute transport during solidification.

Manuscripts resulting from this work:


Phase-Field Models – The phase-field model is attractive for numerical simulations since it al
allows one to solve a free-boundary problem without tracking the location of the interface. Hence, numerical calculations using the phase-field approach are able to successfully simulate anisotropic dendritic growth. However, it is difficult to quantitatively assess these efforts because of the complicated nature of dendritic solidification. Hence, to establish confidence in the phase-field approach workers through rigorous analysis and thorough asymptotic investigations have demonstrated convergence properties of the phase-field equations to the sharp-interface equations as the interface thickness approaches zero. Further, comparisons between numerical phase-field simulations and analytical and numerical sharp-interface results for planar front and needle crystal calculations have shown excellent agreement.

Recent phase field-investigations have focused on selection of the potential coupling the order parameter and temperature. It has been shown that different potentials may lead to different numeric results. Further, by properly selecting parameters in the potential definition, one can enhance the accuracy, in terms of comparison to the sharp-interface model, of the phase-field method.

We conducted further asymptotic investigation of the phase-field equations for this same purpose. Current asymptotic investigations of the phase-field equations have demonstrated that the sharp-interface equations and boundary conditions are recovered at leading order, and have derived estimates on the magnitude of higher order corrections. Generally, no attempts are made to solve the derived full-form sharp-interface equations because this is what one is trying to avoid in the first place. We conducted asymptotic investigations of the phase-field equations that lead to simpler problems at each order. This is accomplished by combining the sharp-interface limit traditionally used to analyze phase-field equations with the asymptotic limits described above which are used to analyze sharp-interface equations. This approach enabled us to develop approximate analytical solutions to the phase-field equations. The leading order approximation of these solutions is a simple sharp-interface solution, and subsequent correction terms are due to a non-zero interface thickness.

We have used this solution strategy to examine the one-dimensional directional solidification of a pure material for the purpose of analyzing the relationship between the solution resulting from a phase-field model to that from a sharp-interface model. An asymptotic analysis based upon a large Stefan number is performed on the sharp-interface model. In the phase-field case, the large Stefan number expansion is coupled with a small-interface-thickness boundary-layer expansion. The results show agreement at leading order between the two models for the location of the solidification front and the temperature profiles in the solid and liquid phases. However, due to the non-zero interface thickness in the phase-field model, corrections to the sharp-interface location and temperature profiles develop. These corrections result from the conduction of latent heat over the diffuse interface. The magnitude of these corrections increases with the speed of the front due to the cor-
responding increase in the release of latent heat. By properly selecting the potential coupling the
order parameter and temperature in the phase-field model, and by tuning the kinetic parameter, we
are able to eliminate the corrections to the outer temperature profiles in the solid and liquid phases
of the phase-field model. This in turn eliminates the correction to the location of the solidification
front. Hence, the phase-field temperature profiles agree with the sharp-interface profiles, except
near the solidification front, where there is smoothing over the diffuse interface and no jump in the
temperature gradients.

We also finished an extension of the above model to include concentration. The sharp-interface
result is the leading order solution discussed in the time-dependent solidification models section,
listed above. Once again we are able to show by proper selection of the kinetic parameter that the
phase-field temperature and concentration profiles agree with the sharp-interface profiles, except
near the solidification front, where there is smoothing over the diffuse interface and no jump in the
temperature gradients and no jump in the concentration profiles. We verified the above findings
by performing numerical simulations of the phase-field equations and comparing those with the
asymptotic results. Further, we began to extend the asymptotic analysis to investigate morphologi-
ical stability using the phase-field approach.

Manuscripts resulting from this work:

Master's Thesis - Lance Nelson, Simulation of a one-dimensional phase-field model for solidifi-


*Scanned Probe Oxidation Model* – Scanned probe oxidation lithography is based on local surface
oxidation induced by an electrically biased atomic force microscope (AFM) tip. The AFM, as shown
in Figure 1, is constructed from a cone-shaped tip that is brought close to or into direct contact
with a substrate. The probe is attached to the end of a cantilever that bends as the tip moves
across the substrate's surface. The deflection of the tip is measured by reflecting a laser beam off
the top of the cantilever. As such, the AFM can detect variations in surface topography. Finally,
by increasing the voltage on the tip, the tip becomes an ion source. Thus it is possible to write
on the substrate as follows. A biased AFM tip in close proximity to a substrate establishes an
intense electric field distribution. As shown in Figure 2, water from ambient air condenses in the
narrow gap capillary between the AFM tip and the substrate to form a liquid bridge. The free
surface of the water droplet that is formed contacts the AFM tip and edge of the oxide or substrate,
depending on the hydrophobicity of the materials. Oxyanions OH\textsuperscript{-} are produced within the water and directed by the electric-field distribution within this fluid region; these ions react with the substrate to produce a thin oxide layer.

Hence, the AFM tip acts as a pen writing oxide lines (see Figure 3) on the substrate. Such oxide patterns have been employed successfully as an etch mask for wet and dry processes, for device isolation, tunnel barrier formation, and as chemical and biological templates. Since most metals, semiconductors, and even insulating thin films such as silicon nitride can be oxidized under such conditions, scanned probe oxidation is a very general method for prototyping nanoscale masks, templates, and devices. Hence, due to the generality of oxidation as a chemical process, the simplicity of the tools and techniques required for producing nanometer-sized features, and the compatibility of the oxidation process with the techniques used by the semiconductor industry, scanned probe microscope (SPM) oxidation using an atomic force microscope tip is a valid approach to nanostructure fabrication.

By understanding the properties of this nanoscale growth it may be possible to build high-aspect ratio (height to width) functional nanostructures. These structures, when coated with application-specific materials, make it feasible to produce nanoscale capacitance-based sensor ele-
Figure 3: A pattern of oxide lines (about 100 nm apart) written on a ZrN surface.

ments. Moreover, by building several such structures on a single substrate and using a variety of coatings, it should be possible to design a single nanoscale multi-sensor capable, for example, of measuring acoustic, thermal, chemical and biological phenomena.

The major limiting factors in constructing these devices are: (1) the ability to consistently build (write) high-aspect ratio structures and (2) the slow speed of the scanned probe writing process. To alleviate these limiting factors and take full advantage of the novel capabilities offered by this nanoscale deposition process, the mechanisms of nanodeposition under varying conditions must be understood across length scales ranging from the atomic to the macroscopic.

We began the development of fundamental mathematical tools to enhance the scientific understanding needed for this critical deposition technology. The central goal of this project is to couple mathematical modeling and analysis to study lithographic patterning techniques based on scanned probe oxidation. Deposition on conducting single material substrates was examined. In all cases deposition takes place in a nanoscale regime, but structures that are micrometers in size can be formed during the patterning. At the nanometer length scale the deposition process leads to the formation of a self-sustaining three-dimensional electrochemical nanocell (see Figure 2) that is not present in macroscopic anodic deposition processes. This nanocell is defined within four free boundaries: a fluctuating atomic force microscope (AFM) tip, and the liquid/gas, liquid/oxide, and oxide/substrate interfaces. Further, there are moving contact lines at the junction of the interfaces and at the AFM tip. The complexity of this nanocell system presents significant mathematical challenges for the solution of the model equations and leads to a significant number of problems for analysis.

The proposed project developed, solved, and analyzed a model for one-dimensional scanned probe oxidation and developed axisymmetric models as a means for writing oxide patterns on a substrate. The model included phase boundaries, reaction-diffusion processes, electric fields, and thermal and mechanical oscillations within the system. Free boundaries separated layers of substrate, oxide and fluid regions. The coupling between the layers influences the mechanical and
reactive mechanisms, ion and electron transport and electromagnetic fields in the system. This type of surface chemistry is most appropriately termed hyper-thermal in that it involves electronically excited neutral and ionic species that do not necessarily follow thermal reaction channels. The models developed for this process broaden the mathematical formalism needed to address similar types of systems. Further these models will be used to increase the uniformity of nano-patterning and will ultimately be applied in process control strategies. We note that this work is ongoing.

Manuscripts resulting from this work:

Master's Thesis - Aaron Orians - One-dimensional growth of an AFM written oxide dot. August 2004

Master's Thesis - Sandra Djurkovic - An asymptotic model for axisymmetric scanned probe oxidation. May 2004


Nanofiber Coating Model – Nanotubes have attracted great academic and industrial interest in recent years. Improvement in the ability to synthesize nanotubes of different materials has resulted in the suggestion and development of novel devices based on the properties of the nanotubes. Possible applications for nanotubes in the areas of filtration, composites, biomedicine, and electronics have been suggested. Carbon nanotubes have also been used to modify atomic force microscope (AFM) tips for nanolithography applications. However, several limitations to the widespread synthesis and use of nanotubes can be identified. First, the ability to produce large quantities of nanotubes with controlled electronic and structural properties is still undeveloped. Second, the nanoscale dimensions of these materials often lead to previously unobserved properties that need to be understood and ultimately controlled. For example, unique conducting properties have been observed in carbon nanotubes. Identifying and understanding these properties is crucial for device design.

This project addressed some aspects of these issues. First, from the standpoint of nanotube synthesis, we to examined physical vapor deposition techniques for applying conductive coatings to nanofibers. Removing the nanofiber core leaves a polycrystalline nanotube of the coating material. Second, from the behavioral standpoint, we examined the electromagnetic response of coated nanofibers and nanotubes. This is relevant due to interest in the electric field emission properties of nanotubes. Nanotube emission characteristics are found to be far better than other electron emit-
ters. Hence, nanotubes offer promising applications as emitters in flat panel displays and microwave power amplifiers. However, there is still little understanding of the fundamental physics of electron field emission from nanotubes. This project examined this issue by modeling the electromagnetic behavior of the coated nanofibers and nanotubes using a combination of QM/MD techniques with continuum equations.

The practice of coating nanofibers is relatively new. Hence, we shall limit our description of experimental observations to those witnessed in the laboratory of Professor Edward Evans, Department of Chemical Engineering, The University of Akron. The nanotubes we are considering are manufactured in a three-step in-house process. First, a polymer nanofiber is created. Electrospinning provides a straightforward method to produce fibers with diameters in the nanometer range. The scientific basis for the mechanism of fiber formation, and control of their physical properties, has already been developed and will not be elaborated upon here. Second, the fibers are coated by plasma deposition methods to produce a core-clad system. Professor Evans has successfully coated fibers with carbon, copper, and aluminum films by using a plasma enhanced physical vapor deposition (PEPVD) sputtering process. Finally, the polymer core (hereafter referred to as the sacrificial fiber) is removed by dissolution, leaving a nanotube of the cladding material. In this way, sacrificial polymer fibers (templates) can be used to create free-standing nanotubes of many materials such as metals and semiconductors. However, a process for synthesizing nanotubes of any length with controlled electric or chemical properties is still required.

Figure 4 shows an aluminum-coated fiber. The cylindrical cross-section of a tube is shown in Figure 5, which indicates that the tube did not collapse after the polymer inside had been removed. The smallest inner diameter of the tubes was around 20 nm. The approximate thickness of the wall of the tubes was controlled by the sputtering process. A tube with different wall thickness is shown in Figure 6.

The above approach can be used to produce tubes of many materials including metals, semiconductors, ceramics and polymers with controlled diameters and a range of nanometer thickness walls. Control over the diameter, thickness, and surface morphology of a nanotube is important.
because of the perceived effect that the wall size and morphology have on the electrical properties of the tube.

Some of the fundamental questions regarding nanotube synthesis and processing, as well as thin film coating in general, are:

- How can one obtain a uniform coating thickness around the outside diameter of the sacrificial fiber?
- In plasma deposition processes, a sheath region forms near the substrate. The ions accelerate through the sheath and bombard the substrate. How do the fiber diameter and the sheath thickness affect the coating uniformity?
- How does the roughness of the coating depend upon the reactor process variables?
- How does the coating thickness and roughness affect the electromagnetic properties of the tube?

The intent of this project’s modeling and simulation effort of nanofiber deposition and the subsequent behavior of coated nanofibers and nanotubes is to make progress toward answering these questions.

To date we have provided fundamental mathematical tools and scientific understanding needed for 1) the investigation of the coating of polymer nanofibers with conducting materials and 2) their subsequent response to applied electromagnetic fields. Experimental procedures for producing the coated nanofibers have recently been developed. However, the interrelationships among processing factors has not been investigated from a detailed modeling approach. Hence, comprehensive models for describing the salient physical and chemical phenomena are needed. The intent of this project was to develop such models across the multiple length and time scales that characterize nanoscale growth.

In the first project area, we began the examination of physical vapor deposition techniques for applying conductive coatings to nanofibers. The modeling focused on the coating growth, its uniformity, morphology and properties. We developed, and are now solving, and analyzing models governing a comprehensive description of the deposition process and performing simulations of in-house experiments. The goals are to explain experimental observations that are not yet well understood and identify the range of parameters that is optimal for nanoscale growth. We are developing solution strategies that couple continuum and atomistic models. At the continuum scale we are developing the appropriate governing equations and boundary conditions to describe the reactor dynamics and deposition of the coatings on the nanofibers (specific to our physical vapor deposition (PVD) reactor environment). We are also developing the governing equations and boundary conditions to describe the electromagnetic fields around a coated fiber. We are using
asymptotic solution methodologies that take advantage of the disparity in length and time scales within these systems to reduce the models to tractable forms. The reason for this solution approach is that we plan to couple continuum models with atomistic simulations. Hence, we shall use the asymptotic approach to seek a compromise between retaining physics from the reactor scale level and keeping the numerical simulations to a manageable level. From the asymptotic simulations, the normal growth velocity is being used for level set method simulations, which numerically calculates the location of the coating front at the continuum length scale. We are testing the validity of the continuum models through comparison with experiments.

At the atomic level, we are beginning to use quantum mechanical (QM) and molecular dynamics (MD) simulations. We are studying properties of the deposition mechanisms, the nanofiber coating, migration of atoms in the coating, and calculating electronic properties of the resulting nanotube (after dissolving the inner core) as a function of the coating thickness.

The continuum and QM/MD simulations complement each other. Information about ion and neutral species energy and flux, and the thickness of the coating is being passed to the QM/MD models from the continuum equations that describe the coating growth. The QM/MD models are being used to integrate across length scales to calculate deposition properties and electronic properties of a metallic coated nanofiber as a function of the coating thickness. These properties are then carried to the continuum model to study the overall electromagnetic fields about the resulting nanotube. The idea is to determine the coating parameters which yield desired electronic properties, for applications such as nano-emitters.

The model for the coating of nanofibers is based upon deposition within a traditional PEPVD system. Our objective is to determine the influence of process conditions on the uniformity and morphology of the coating. A traditional PEPVD sputtering system is characterized by a bulk gas phase dominated by neutral species, and sheath regions that separate the bulk gas phase from the substrate (nanofibers) and the target, as shown in Figure 7. The potential is greater in the plasma region than in either sheath region. Hence, gas ion molecules (e.g. argon) accelerate toward and bombard the target material (e.g. aluminum) and sputter neutral species away from the target source. Based upon the distance from the target to the substrate, the sputtered material either ballistically impacts (short distances) the substrate, or diffuses (far distances) toward the substrate. For simplification, we assume that only the neutral species deposit to form the coating. However, the ions will also accelerate to the substrate and sputter material from the deposited coating. This material re-emits into the plasma. Hence, the coating process is complicated by the sensitivity to both the reactor scale process variables and the molecular topography of the coating. This integrated feedback deals with different length scales, spanning tens of centimeters at the reactor scale to nanometers at the molecular feature scale of the coating. These factors indicate a strong need for an integrated approach in the simulation process, in which both the reactor
properties and the details of the topography will be taken into account. The reactor level process variables influence the molecular feature coating profile, while the coating feature processes can affect the entire reactor level characteristics as feature scale information is introduced to the global level. Simulation of this feedback between molecular features and the reactor level is difficult to accomplish given current computational abilities. Thus, efficient solution methodologies integrating simulations at the various length scales must be developed. The descriptions above present our strategy for accomplishing this integration through the linking of models at the global reactor scale, the local nanofiber scale, and the molecular scale.

The model for coating deposition includes the dynamics of the bulk gas phase transport of neutral molecules, the electromagnetics that govern the ion transport through the sheath regions, and the interaction/reaction of the ions with the substrate. By combining a surface deposition model using QM/MD simulations at the fiber surface with models for the plasma, we obtain a comprehensive model of the entire reactor system across atomic to macroscopic scales. We note that this work is ongoing.

Manuscripts resulting from this work:

Master’s Thesis - Toma Marinov - An asymptotic model for field emission around a nanowire. August 2004

Senior Honors Project - Ines Busuladzic - Formulation and solution of models for the sheath region in a plasma. May 2004

6 Personnel

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