Title: **Industrial Processing of Complex Fluids: Formulation and Modeling**

Author(s): J. C. Scovel, CIC-3
S. Bleasdale, CIC-3
G. M. Forest, Eng. Mech. Dept., Ohio State U.
S. Bechtel, Eng. Mech. Dept., Ohio State U.

Submitted to: DOE Office of Scientific and Technican Information (OSTI)

DISCLAIMER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

Los Alamos
NATIONAL LABORATORY

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Industrial Processing of Complex Fluids: Formulation and Modeling

James C. Scovel* and Shirley Bleasdale
Computing, Information, and Communications Division, Los Alamos National Laboratory

Greg M. Forest and Steve Bechtel
Engineering Mechanics Department, Ohio State University

Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The production of many important commercial materials involves the evolution of a complex fluid through a cooling phase into a hardened product. Textile fibers, high-strength fibers such as KEVLAR and VECTRAN, plastics, chopped-fiber compounds, and fiber optical cable are but a few examples of such materials. Industry contacts for each of these materials are keenly aware of the physics and chemistry that dominate their manufacturing processes and desire to replace experiments with on-line, real time models of these processes. Industry scientists are equally aware of a humbling fact: solutions to their problems are not just a matter of technology transfer, but require a fundamental description and simulation of their processes that lies just beyond the current state of science. The goals of our project are to develop models that can be used to optimize macroscopic properties of the solid product, to identify sources of undesirable defects, and to seek boundary-temperature and flow-and-material controls to optimize desired properties.

Background and Research Objectives

The important elements of these material processes consist of a complex fluid, usually with significant non-Newtonian rheology, temperature dependent viscosity, thermal variations from liquid to solid phase, and the most elusive and least understood orientation effects at particular length scales (molecular scales in KEVLAR type materials, intermediate or mesoscales in many textile fibers and plastics, and macroscales in chopped-fiber compounds) which couple to the thermal flow and solidification process. Internal length-scale orientation of the finished product dominates the desired properties, and yet this is the weakest link from the basic science perspective. We note the commonality of this multiple length-scale coupling to various materials processing problems addressed by others at Los Alamos.

As a result of the complexities of these systems, significant compromises are made to achieve the existing crude models which fall short of their full potential—to troubleshoot existing processes and materials, and to perform parameter studies for the design of new

---

* Principal Investigator, E-mail: jcs@lanl.gov

materials and processes. For example, the Hoechst-Celanese Corporation uses a fiber spinning model which ignores polymer orientation in the flow and then applies empirical relations to infer orientation from the computed stress field. This orientation information is then used to predict tensile strength and optical properties of the fiber. This is only one of the many features absent: transient dynamics and stability information, significant gradients transverse to the fiber axis in temperature are presumed zero, etc.

Clearly, a coupled thermal-flow-orientation-solidification model of this free surface flow (and related problems) is both unavailable and highly desirable. The goal of this project is to develop the capability to predict, for example, the orientation/stress field relationship as a function of model parameters. Such high-level models can be used to optimize macroscopic properties of the solid product, to identify sources of undesirable defects, and to seek boundary, temperature, flow and material controls to optimize desired properties.

Importance to LANL's Science and Technology Base and National R&D Needs

The coupling of various length-scale orientation effects to flows is itself a critical basic scientific problem. The added effects of temperature dependence and phase change to solidification, with free surfaces, pose an opportunity to advance fundamental science and simultaneously assist US industry in gaining a competitive advantage.

Furthermore, these capabilities may contribute to and gain from other industrial and basic material science efforts here at LANL concerning the processing of metals and ceramics. Although the specific physics and engineering of metals and ceramics are different, at the fundamental level they are remarkably similar: internal length-scale structures (orientation effects) couple to the macroscopic length scales and critically determine the desired macroscopic properties such as strength, defects, and post-processing deformation.

For example, in the laser welding project at Los Alamos of Tony Rollet (MST-6) and Richard LeSar (CMS), they must model the melting, flow, deformation and re-solidification of the metal, which is fundamentally influenced by the microstructure. The technical and technological impact of accurate, flexible processing codes could be dramatic. As textile and optical cable industry contacts have noted, a 5-10% gain in product efficiency translates to market domination. Demonstration of such capabilities should encourage industry to engage in CRADAs to collaborate towards the development of on-line codes to aid in the industrial production of these advanced materials.

Scientific Approach and Accomplishments

Initially, we began by concentrating on the mold filling and solidification process. In particular a highly viscous melt is forced into a mold and simultaneously cooled to obtain a solid with prescribed shape. Modeling this process amounts to understanding how to model and simulate solidification in the presence of slow flow in a prescribed domain. We have isolated the areas that must be understood individually and then coupled.

1. Flow of the molten polymer.
2. Heat transfer in the melt domain.
3. Interface mechanics.
4. Solidification of the melt.
5. Heat transfer in the solid domain.
6. Thermomechanical stress and deformation analysis of the solid.

Each of the above areas will constitute a module in a driver code.

- We have searched for a finite element fluid code that can handle phase changes and non-Newtonian fluids. We have found that the code FIDAP appears to suit our needs.

- We have successfully implemented the fluid code FIDAP in two-dimensional geometry on a quasi-steady state solidification problem in simple geometry.

- We began deriving equations coupling the orientation effects based on Ericksen's liquid crystal models. These equations can be simplified to standard form and at the same time give qualitative theoretical characterizations of observed physical phenomena.

We derived a multiscale formulation of complex fluids based on a nonlinear state space model where the observation equation corresponds to the relation between the small scales and the larger scale structures. Significant information is available regarding the solutions of such models for linear systems through application of the Kalman filter. Consequently, we reduced the scope to linear systems. However, even then our researches showed that little is known about multiscale state space models. Consequently, we concentrated on the relevant elementary unsolved problem: how to disaggregate linear time series data.

We developed and tested a disaggregation procedure for time series data based on an EM (expectation and maximization) type algorithm we derived. However, testing this algorithm on ARMA (auto regressive moving average) time series models gave mixed results

3

until we discovered the work of Al-Osh who described the likelihood function for the method [1]. Our techniques combined with maximizing the likelihood function gave very interesting results as follows:

- The parameters of the ARMA models generating the original data can be well determined.

- The actual data is not particularly well determined by this method when compared to a naive disaggregation technique at a pointwise level. However, upon computing autocorrelation functions, it is observed that our technique produces data that is more like data from a stochastic process of the correct type.

- We produced an extensive investigation into the behavior of our algorithm in the specific parameter regimes of AR, MA, and ARMA time series models with interesting results.

- The relevance of these results to complex fluids is that if the pointwise stationary data is all that is important, more naive approaches for multiscale descriptions might be relevant. However, since the relaxational modes of a complex fluid are related to how the fluid is evolving and how the different scales are interacting, such a technique shows promise. The next step in such an evaluation is the development of a nonlinear analogue of the Kalman filter.

Publication
1.	Bleasdale, S., Burr, T., and J. Scovel, "Disaggregating Time Series Data," Los Alamos National Laboratory report, in preparation.

Reference
[1]	Al-Osh, M., "A Dynamic Linear Model Approach for Disaggregating Time Series Data," Journal of Forecasting, 8 (2), 85–96 (1989).