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SANTA BARBARA • SANTA CRUZ

(Handwritten initials)

DEPARTMENT OF CHEMICAL AND
NUCLEAR ENGINEERING
TELEPHONE: (805) 893-3412
FAX: (805) 893-4731

SANTA BARBARA, CALIFORNIA 93106-5080

AD-A267 020



January 9, 1992

Mr. Clint Warner
Office of Naval Research
565 South Wilson
Pasadena, CA 91106

Re: Final Report ONR 25-784811

Dear Mr. Warner,

Enclosed is the final report (at long last!) which we discussed in a telephone conversation on October 31, 1991. Please understand that the delay from that point in time was caused by my procrastination of the final preparation of the manuscript and not Gary's lack of attention to the matter.

Very truly yours,

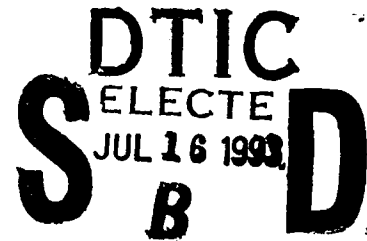
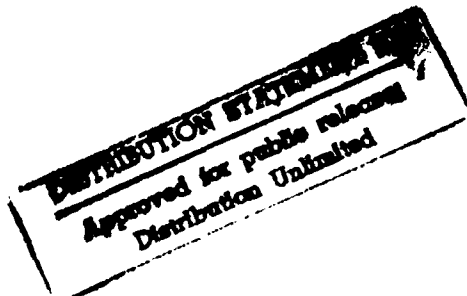
Bonnie C. Franzen

Bonnie C. Franzen
Assistant to L. Gary Leal

Enclosure

cc: Earl Friese
UCSB Office of Research

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FINAL REPORT
ONR 25-784811
"Macromolecular Flows"

I. INTRODUCTION

One of the unresolved problems of fluid mechanics with the greatest potential for important applications is turbulent drag reduction induced via the addition of additives, usually minute quantities of very high molecular weight polymer. This problem is unresolved in the sense that the basic mechanism remains a matter of controversy, and thus, the ability to optimize the process is limited. In part, this is because of the complexity of the base flow--i.e., turbulent flow near a solid boundary--and a lack of knowledge about the structure and dynamics of this flow in the *absence* of polymer. Equally, however, it is because of an incomplete understanding of the dynamical properties of dilute polymer solutions. The quest to understand (*predict*) drag reduction, in either qualitative or quantitative terms, holds the potential of pushing forward the frontiers in both areas--if we can understand *in general* how minute quantities of polymer can influence fluid motions, this may provide vital clues to understanding not only drag reduction, but also the underlying turbulence. Likewise, understanding turbulence and the way that it may be influenced by additive, can yield important insight into the dynamics of dilute polymer solutions.

We have been involved in a long-term study, based upon ONR support, that is aimed primarily at the behavior of dilute polymer solutions. A comprehensive summary of this work was presented as one of the keynote invited lectures at the recent IUTAM Symposium on Drag Reduction in Zurich¹ (presented by Professor Sanjoy Banerjee). A written version of the talk has been published and is attached to this report.

II. SUMMARY OF RESEARCH PROGRESS

Previous studies in this lab²⁻⁹ were focused on the behavior of dilute (well-mixed) solutions of well-characterized polymer (known molecular weight distributions, etc.) in relatively simple, steady extensional flows. These studies were initially aimed at understanding and

predicting the transition from a coiled to a stretched configuration in a spectrum of 2D strong flows ranging from pure extension to a very close approximation of simple shear flow. More recently,¹⁰⁻¹² we have obtained detailed measurements of *the changes in the flow field which occur as a consequence of this polymer conformation transition*. Besides giving detailed data that can ultimately be compared with theoretical predictions, these studies suggest that the coil-stretch transition process must lead to a high degree of *polymer-polymer interaction* if the polymer in a dilute solution is to have any effect on the flow. Further, the effect in steady, laminar flows is *very highly localized*, corresponding to a decrease in strain-rate in a very narrow region along the outflow axis that begins at the hyperbolic stagnation point of the flow.

In the most recent grant period, we initiated a number of projects aimed at the next step, which is to obtain quantitative theoretical predictions for the non-trivial flows as a preliminary to attempts at numerical simulation of turbulent drag reduction. We also began to consider experiments, models for the polymer, and predictions for flows that are closer to turbulence. We shall discuss the latter topics shortly. First, however, let us return to the ongoing efforts to obtain predictions for the flows that we have already studied experimentally, beginning with a re-statement of the motivation and goals for this part of our work.

In particular, it is important to recognize what we do know and what we do not know about current theoretical models for the polymer and for the bulk flow properties of dilute solutions. As long as we restrict ourselves to steady, strong flows, it is fair to say that the preceding work has led to theoretical models that very successfully describe the *conformation* of the polymer in a *known* velocity field, including the complete transition from a random coil to a fully stretched and oriented state. The restriction to (Eulerian) steady flows is primarily a limitation on the time-dependent flow history (Lagrangian) that is seen by the polymer. Basically, existing *predictions of polymer conformation* work well in flows, which from a Lagrangian point of view, present monotonic variations in strain-rate with no abrupt changes of either magnitude or flow type. It is important to recognize, however, that there is *no* assurance that the existing models will provide a reasonable *prediction of the changes in the flow* (relative to the Newtonian fluid) that are caused by

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the polymer. Indeed, our experiments¹⁰⁻¹² have shown that the polymer only begins to influence the flow in a *dilute* solution if it is sufficiently stretched to yield strong and extensive polymer-polymer *interactions*, and the existing dilute solution models^{8,13-15} do not take polymer interreactions into account at all--either in the polymer contribution to the bulk stress, *or* in the dependence of the polymer conformation on the flow. The only way to test existing models and assess their deficiencies from a bulk fluid mechanics point of view, is to compare numerical predictions of both the polymer conformation and the flow field with experimental data in flows where there is a significant *measurable* effect of the polymer on the flow, and a baseline of experimental data. *To date, this has not been done for any single flow!* The problem is two-fold: first, except for our recent measurements there is not a single set of experimental data with simultaneous measurement of polymer conformation *and flow*; secondly, the solution of the full fluid mechanics problem requires simultaneous *numerical solution* of the equations of motion and the coupled nonlinear (dumb-bell) model for the polymer and this has proven to be extremely difficult (though we are very close to achieving that goal--see A-1 below).

A. *QUANTITATIVE THEORETICAL PREDICTIONS OF FLOWS THAT ARE STEADY FROM AN EULERIAN POINT OF VIEW*

A summary of *recent* progress related to the goal of *predicting* the flow behavior in steady, strong flows is as follows:

1) *NUMERICAL CALCULATIONS FOR THE MOTION OF A DILUTE SOLUTION OF DUMBBELLS IN A TWO-ROLL MILL*

We are currently in the midst of a major effort to obtain numerical solutions for the motion of a dilute polymer solution, modelled via the nonlinear dumbbell with conformation-dependent friction, in a two-roll mill with the same geometry as used in previous experimental studies from our laboratory.

The chief difficulties with this numerical problem are two-fold: First, the "minor" problem is to deal with the complex geometry of the experimental two-roll mill (two

rotating cylinders in a box); second, there is the *intrinsic* difficulty of obtaining numerical solutions for non-Newtonian fluids. The first problem has long-since been resolved, and a paper is being written to report on the interesting fluid mechanics of a *Newtonian* fluid in both steady and time-dependent versions of the two-roll mill. The second problem is, in some sense, generic to all efforts to obtain numerical solutions for viscoelastic flow problems (the so-called "high Deborah number problem")¹⁶ and is manifested by the appearance of regions within the flow domain (i.e., *not* usually at the boundaries) where the stress and stress gradients become *very large*. This is exacerbated in the dilute solution case by the fact that the changes in polymer conformation (and in fact the whole interaction between the polymer and flow) are *extremely localized*. Modest success in obtaining numerical solutions has been achieved for constitutive models that are intended to apply to *concentrated* systems by either introducing "ad hoc" modifications in the models to smooth large stress gradients,¹⁷ or by using multi-grid systems with a much greater spatial resolution of stress than of velocity.¹⁸ In the present case of dilute solutions, we are loathe to introduce "non-physical" modifications of the models, and have been working primarily via enhanced spatial resolution for the representation of polymer conformation (i.e., for the second-moment tensor) $\langle \mathbf{r} \mathbf{r} \rangle$, where \mathbf{r} is the end-to-end vector, as in other recent work.¹⁹ Strangely, however, we have also discovered that one of the fundamental assumptions that underlies the classic dumbbell model derivation breaks down in regions of large conformation gradients, and this has also led to the recent development of a fundamentally new version of the basic dumbbell theory¹⁵ (described below) which actually minimizes the presence of these gradients and suggests that their origin (perhaps in *all* viscoelastic model fluids) is *aphysical* (i.e., that our numerical difficulties with non-Newtonian flow calculations reside at least partially *inadequate models*).

(2) IMPROVED POLYMER (CONSTITUTIVE) MODELS FOR DILUTE SOLUTIONS

As indicated above, we have made what we believe to be a very significant breakthrough in the development of model equations for the dynamics (and bulk rheological properties) of dilute polymer solutions. This development had its origin in the numerical work described above. Specifically, in view of the extreme difficulty of obtaining reliable numerical results (and the similar difficulties experienced by *all* major research groups who were trying to compute any kind of non-Newtonian flow), we felt that it would be desirable to establish a rigorous mathematical proof of the conditions for the *existence* of steady solutions of the equations of motion for our dilute polymer model in a bounded domain. With such a proof in hand, it would be possible to distinguish between difficulties of a strictly numerical character, and "difficulties" associated with the lack of existence of steady solutions for our model system (i.e., presumably of non-physical breakdowns of the model).

The interesting thing is that we were able to actually complete *a rigorous proof of existence for steady flows in a bounded domain, for all Deborah numbers*, but only if the dumbbell model was modified to include one term in the microstructure evolution equation that is normally neglected. Further, although this added term was originally suggested strictly on mathematical grounds, a very careful re-evaluation of the derivation of the model¹³ showed that it represented a physical effect that should strictly be present, but is usually neglected as an *approximation* in the model. This effect corresponds to *spatial* diffusion of the polymer which tends to *smooth* gradients in polymer conformation. The classical assumption is that *spatial diffusion* can be neglected (i.e., "local homogeneity") but we find in reality that the relevant term in the model is actually multiplied by a *small* (but nonzero!) *parameter* so that it is negligible in normal circumstances, but becomes significant in regions of large gradients of polymer conformation (i.e., just in those regions where numerical schemes have typically experienced extreme difficulty!) We feel that our discovery of the "missing term" in the dilute solution model is a major breakthrough, which

most likely has profound implications for a much wider class of constitutive equations since these all experience similar numerical difficulties.

Our work on this problem, including both the modified model and the proof of existence, has been published in the *Journal of Non-Newtonian Fluid Mechanics*.¹⁵

(3) POLYMER-POLYMER INTERACTION EFFECT IN DILUTE SOLUTIONS (EXPERIMENTAL)

We have noted above that polymer effects appear in the *motions of dilute* solutions, only when the polymer is sufficiently stretched to induce a high level of polymer-polymer interaction. In the drag reduction context, however, this observation is somewhat puzzling because it is well known that there is a critical concentration of polymer *above* which the degree of drag reduction actually begins to *decrease*.^{20,21} The usual "explanation" for this is that the polymer concentration has been increased to the point where *the shear viscosity* is increased by the polymer, and the *preferential damping of extensional deformations* that is the key property of dilute solutions, begins to be overshadowed by this increase. However, this does not seem to be a very convincing argument, especially in view of the fact that we need a transition to an *effectively non-dilute system (i.e., highly interacting)* to *get any flow effect at all*.

Hence, in the present work, we have carried out a systematic series of experiments to examine the influence of polymer interactions on flow-induced extension in strong flows, including an attempt to compare experimental observations with modified versions of the molecular (dumbbell) models that are intended to account for polymer-polymer interactions. This was done by considering the behavior of polystyrene solutions of MW ~ 2×10^6 at three concentrations - 100 ppm, 1500 ppm and 4500 ppm. The latter two concentrations correspond approximately to $1/3 C^*$ and C^* where C^* is the concentration at

which polymer-polymer interaction first becomes discernable for the polymer in its equilibrium configuration.

The results of this study were initially surprising to us (although, in retrospect, perhaps they should not have been in view of the drag reduction and other results mentioned above). Our measurements showed that the degree of polymer extension (and orientation) was highly *inhibited* by polymer-polymer interactions, including the *final* "asymptotic" level at high strain rates. This, in itself, is not so surprising. However, the magnitude of this inhibition is such that the *total* effect of the polymer on the flow actually *decreases* when the concentration is *increased* from 1500 to 4500 ppm, and this (to us) was totally unexpected. Subsequently, we have investigated a number of theoretical models that are intended to account for particle-particle interactions.^{22,23} Among these, we found that a semi-empirical model due to Hess²³ provided the only predictions that were in qualitative accord with our measurements, including the observed $C^{1/2}$ dependence of birefringence on concentration. This work is currently being submitted for publication.²⁴

B. TIME-DEPENDENT AND HETEROGENEOUS FLOWS

The second major goal of current research has been the initiation of studies that are intended to explore polymer behavior in *transient* flows that provide a more realistic assessment of current models for application to turbulent flows (e.g., via computational simulations). The essential motivation for these studies is the simple question of whether the "molecular" models derived for application to steady *strong* flows, can provide realistic predictions when the variations in the flow are much faster and more complex. An attempt to address this question must ultimately involve a number of complicated and seemingly disparate problems.

(1) *POLYMER MODELS FOR APPLICATION TO HETEROGENEOUS AND TIME-DEPENDENT FLOWS*

First, and perhaps most important, is the necessity to understand the dynamics of a single polymer molecule in heterogeneous and/or time dependent flows. Although this problem might generally be presumed to be solved, all current theories are based upon the *approximation* of such flows via a sequence of *homogeneous* flows in which the velocity gradient tensor at any instant is assumed to be a *constant*.¹³ However, in general, if we account for the fact that the strain-rate and vorticity E and Ω actually depend on time, the usual equation for the dynamics of a stretchable, orientable macromolecule becomes *non-autonomous* and the predicted dynamical behavior will generally be *qualitatively* different from predictions obtained via the homogeneous (linear) flow approximation which leads to an *autonomous* dynamical equation. This is *very* important at the level of individual polymer dynamics. Further, in view of the fact that *all* dilute solution models (as well as all other macromechanical models) are derived via the local homogeneous flow approximation, we expect it to have profound implications for the rheological behavior of dilute solutions in strongly heterogeneous or unsteady flows!

To date, we have considered only the particular case of a general dumbbell model in a two-dimensional flow, for both aperiodic and periodic time-dependence as seen from the viewpoint of the polymer.²⁵ Even in this case, we can show that the *strong-flow* ("coil-stretch") criteria for a *linear* homogeneous flow (c.f., Tanner,²⁷ Olbricht,*et al.*⁶ etc.) will generally give the wrong answer, both qualitatively and quantitatively, when applied to a *heterogeneous* flow. In particular, we have shown by example, that the usual strong-flow criteria can be satisfied *without* the polymer achieving any significant stretching and vice versa. A key question, that we do *not* currently understand, is the implication of non-autonomous behavior at the microscale, on the bulk constitutive models for dilute solutions that are obtained from the microscale dynamics equation by taking moments and averaging.

These questions address deep fundamental issues associated with the application of dilute polymer solution models to flows that are time-dependent from a Lagrangian point of view. In particular, they hold the potential to radically change our understanding of the dynamics of flexible-chain polymer molecules in flows with complex spatial or time-dependent structure. This is precisely the necessary direction for research that is to be aimed at either understanding or numerical simulations of turbulent drag reduction. Two papers describing our findings have been published^(25,26) but we have only truly scratched the surface of this very important problem area.

(2) *EXPERIMENTAL STUDIES OF POLYMER DYNAMICS IN COMPLEX, TIME-DEPENDENT LAMINAR FLOWS*

The second major new initiative is a continuation of objectives outlined several years ago for studies of polymer dynamics in more general time-dependent flows. It is clear, if we are ever to achieve meaningful numerical simulations of the influence of polymer on turbulent flows that we must have a clear knowledge of the polymer model in flows that vary in a much more rapid and complicated way than anything that has been studied to date. There is a strong indication in recent dynamical simulations of bead-spring chains in rapidly varying strong flows²⁸ that the polymer may not stretch affinely, thus contributing strongly to the solution rheology in both the "*fully stretched*" and "*stretching*" modes. However, even if the current model (in a proper non-autonomous form) is accepted as it stands, the application to rapidly varying flows requires that both the stretch and relaxation dynamics be *quantitatively* correct, and this is *not* true of the flows that have been studied to date. For example, the behavior of a polymer molecule in a flow that is alternately "weak" and "strong" will depend critically upon the time scale of the polymer relaxation process as a function of the degree of stretch,²⁹ and to capture this correctly in the usual dumbbell model requires not only that the spring law and the friction-law be qualitatively reasonable, but also that they have the correct dependence in *detail* on the end-

to-end dimension. *None of the comparisons made previously between polymer conformation predicted from the model and measured from experiments is sufficiently sensitive to distinguish these details.*

Current and proposed research includes two distinctive types of study. *First*, during the past several years, we have built a completely new experimental system based upon a computer-controlled two-roll mill, with two-color birefringence and dichroism, and dynamic light scattering,³⁰ to allow measurements of polymer conformation and fluid dynamics (velocity gradients) in a variety of extremely well-controlled *time dependent* strong flows. Achievable flow histories include stop-start, relaxation, double-step up or down, ramps, or more complicated flows such as sinusoidal variations in strain-rate.

Secondly, we have initiated a study of polymer dynamics in a specially constructed two-roll mill, that is designed to allow alternating variations in roller speed, including a *blinking* mode (BTRM) which "simulates" a so-called blinking vortex flow.³¹ The point of this work is to study polymer conformation and flow modification in a laminar, chaotic flow.

III. PUBLICATIONS AND PRESENTATIONS from N00014-87-K-0137

A. Papers

1. "Dilute Polystyrene Solutions in Extensional Flow: Birefringence and Flow Modification," P. N. Dunlap and L. G. Leal, *J. Non-Newtonian Fluid Mechanics* 23, 5 (1987).
2. "An Experimental Study of Dilute Polyelectrolyte Solutions in Strong Flows," P. N. Dunlap, C. H. Wang and L. G. Leal, *J. Polymer Sci., Part B: Polymer Physics* 25, 2211 (1987).
3. "The Existence of Solutions for All Deborah Numbers for Non-Newtonian Fluids," E. El-Kareh and L. G. Leal, *J. of Non-Newtonian Fluid Mech.* 33, 257-287 (1989).
4. "Orthogonal Grid Generation in a 2-D Domain via the Boundary Integral Technique," I. S. Kang and L. G. Leal, *J. Comp Phys.*, accepted (1991).
5. "Rigid Particles Suspended in Time-Dependent Flows: Irregular vs. Regular Motion, Disorder vs. Order" A. J. Szeri, W. J. Milliken and L. G. Leal, *J. Fluid Mechanics*, accepted (1990).
6. "On the Dynamics of Suspended Microstructure in Heterogeneous Fluid Flows," A. J. Szeri, S. Wiggins and L. G. Leal, *J. Fluid Mech.*, 228, 207-242 (1991).
7. "Concentration Effects on Birefringence and Flow Modification of Semi-Dilute Polymer Solutions in Extensional Flows," R. C.-Y. Ng and L. G. Leal, in preparation.
8. "Chaotic Mixing and Transport in a Two-Dimensional Time Periodic Stokes Flow--The Blinking Two-Roll Mill (BTRM): I. Newtonian Fluids," R. C.-Y. Ng, D. F. James and L. G. Leal, in preparation.
9. "Chaotic Mixing and Transport in a Two-Dimensional Time Periodic Stokes Flow--The Blinking Two-Roll Mill (BTRM): II. Dilute Polymer Solutions," R. C.-Y. Ng, D. F. James and L. G. Leal, in preparation
10. "Dynamics of Dilute Polymer Solutions," L. G. Leal, *Proc. IUTAM Symposium on Polymer Drag Reduction*, Zurich, Springer-Verlag, Berlin-Heidelberg (1990).
11. "An Analytic and Numerical Study of Flow in a Two-Roll Mill." T. Kapur, M. Shapira, E. Ascoli and L. G. Leal, in preparation.

B. Theses

1. "I. On a New Constitutive Equation for Non-Newtonian Fluids; II. Brownian Motion with Fluid-Fluid Interfaces", Ardith El-Kareh, March 1989.
2. "Semi-Dilute Polymer Solutions in Strong Flows. Part I: Birefringence and Flow Modification in Extensional Flow. Part II: Chaotic Mixing in Time Periodic Flow," Ricky Chiu-Yin Ng, October 1989.
3. "Birefringence of Polymer Solutions in Time-Dependent Flows," Enrique Geffroy-Aguilar, November 1989

C. Talks/Presentations

1. International Conference on Extensional Flow, Chamonix, France, January 1988.
2. International Conference on Extensional Flow, Combloux, France, March 1989.
3. Dynamics of Concentrated Systems, Los Alamos National Laboratory, Los Alamos, New Mexico, June 1989.
4. IUTAM Symposium on Turbulent Drag Reduction, Zurich, July 1989 (paper read by S. Banerjee)
5. Session on Optical Methods for Structure and Dynamics of Polymeric Materials, Annual Meeting, AIChE, San Francisco, November 1989.
6. IUTAM Symposium on Fluid Mechanics of Stirring and Mixing, La Jolla, California, August 1990.

IV. REFERENCES

1. "Dynamics of Dilute Polymer Solutions," L. G. Leal, *Proc. IUTAM Symposium on Polymer Drag Reduction*, Zurich, Springer-Verlag, Berlin-Heidelberg (1990).
2. "Studies of the Flow-Induced Stretching of a Macromolecule in a Dilute Solution," L. G. Leal, G. G. Fuller and W. L. Olbricht, *Prog. in Astronautics and Aeronautics* 72, 351 (1980).
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5. "The Effects of Conformation-Dependent Friction and Internal Viscosity on the Dynamics of the Nonlinear Dumbbell Model for a Dilute Polymer Solution," G. G. Fuller and L. G. Leal, *J. Non-Newtonian Fluid Mechanics* 8, 271 (1981).
6. "Strong Flow Criteria Based on Microstructure Deformation," W. L. Olbricht, J. M. Rallison and L. G. Leal, *J. Non-Newtonian Fluid Mechanics* 10, 291 (1982).
7. "The Charged Dumbbell Model for Dilute Polyelectrolyte Solutions in Strong Flows," P. N. Dunlap and L. G. Leal, *Rheol. Acta* 23, 238 (1984).
8. "A Study of Conformation-Dependent Friction in a Dumbbell Model for Dilute Polymer Solutions," N. Phan-Thien, O. Manero and L. G. Leal, *Rheol. Acta* 23, 151 (1984).
9. "Birefringence Studies of Flow-Induced Conformation Changes in Polymer Solutions," L. G. Leal, Proc. Int. Congress of Rheology, Vol. 1, 191 (1984).
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34. "An Analytic and Numerical Study of Flow in a Two-Roll Mill," T. Kapur, M. Shapira, E. Ascoli and L. G. Leal, in preparation.