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FINAL REPORT

COMPARATIVE EFFECTS OF DRAG-REDUCING POLYMERS

AN EXPERIMENTAL REPORT

Prepared for:
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Submitted by:
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13. ABSTRACT (Maximum 200 words)

The experiments described in this report were performed within the framework of the overall ASTI Team effort in polymer science. The underlying predicate of this program has been to achieve a basis for understanding the shear viscosity of dilute polymer solutions through a detailed description of the molecular interactions. Interaction descriptions must include water/polymer, water/surface and polymer/surface contributions to provide understanding of the mechanisms determining the shear viscosity and related noise and drag reduction effects.

Although certain anionic polymeric drag reduction effects observed herein are not dramatic compared to those achieved with the standard polyethylene-oxide, these types of polymers offer the potential for drastically altering the interaction between the surface and adjacent layers of water molecules. It can be assumed that the water layer/surface interaction is dependent on the nature of the surface potential and the presence of strongly interacting species in that region. Thus, any mechanism reducing the concentration of such active species near the surface should reduce the overall strength of the interaction. However, drag reduction actually increased for polyacrylamide changing from deionized water to "instant ocean" (salt water environment). This suggests the role of ions in determining polymer effectiveness may be complex.

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LIST OF SYMBOLS

R_G = Ion Gyro Radius

Δ_{BL} = Boundary Layer Distance

L_p = Polymer Unit Length (average)

d = Interpolymer separation

R_i = Ion-H_2O, Ion-Surface Interaction Distance

R_M = Molecular Interaction Distance

R = Radius of Rotating Disk in Drag Tester

Ω = Angular Velocity

ρ = Density of the Fluid (corrected for temperature)

μ = Viscosity of the Fluid (corrected for temperature)

T = Torque Required to Rotate Disk

A = Surface Area of the Disk (both sides)

$$K = \frac{\rho R^2 \Omega^2}{2} = \frac{\rho u^2}{2}$$

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PREFACE

The experiments described in this report were performed at USBI by Ms. Gail Murphree, principal investigator, at CORTANA's direction under Task II.B: Traveling Wave, Boundary Layer and Vortex Control. It is important to discuss the experimental shear viscosity results of USBI in the framework of the overall CORTANA ASTI Team effort in polymer science. The underlying predicate of this applied research program has been to achieve a basis for understanding of the shear viscosity of dilute polymer solutions through a detailed description of the molecular interactions. Such interaction descriptions must include water/polymer, water/surface and polymer/surface contributions to provide a detailed understanding of the mechanisms which determine the shear viscosity and related noise and drag reduction effects. The phenomenological models advanced thus far neither explain drag reduction effects nor provide a predictive basis for noise reduction phenomena in such systems.

A primary postulate of the ASTI Team approach is that the *interaction* between the first several water layers and the surface is critical in determining both the history and nature of the boundary layer growth and associated transition phenomena. Clearly, the nature of this interaction determines the friction at the surface, and the polymer can be instrumental in perturbing (weakening) the water molecule/surface interaction. The wide range of characteristic lengths of interest for dilute polymers in salt water, in the presence of a magnetic field, is illustrated in Figure 1 below. In this regard it is important to note Ms. Murphree's observation that better drag reduction might have been achieved with selective use of high molecular weight segments. This dependence on molecular weight has been observed experimentally; however, the CORTANA Team would caution that the role of other molecular factors may be masked by such a simplistic interpretation of data. These other molecular factors include chain length, radius of gyration, and hydrodynamic volume.

Although certain anionic polymeric drag reduction effects observed herein are not dramatic compared to those achieved with the standard polyethylene-oxide

(PEO), these types of polymers do offer the potential for drastically altering the interaction between the surface and adjacent layers of water molecules. It is plausible to assume that the water layer/surface interaction is dependent on the nature of the surface potential and presence of strongly interacting species in that region. Thus, any mechanism which reduces the concentration of such active species near the surface should reduce the overall strength of the interaction. It is not, however, encouraging to note that drag reduction actually increased for polyacrylamide in going from deionized water to "instant ocean" (salt water environment). This suggests the role of ions in determining polymer effectiveness may well be quite complex.

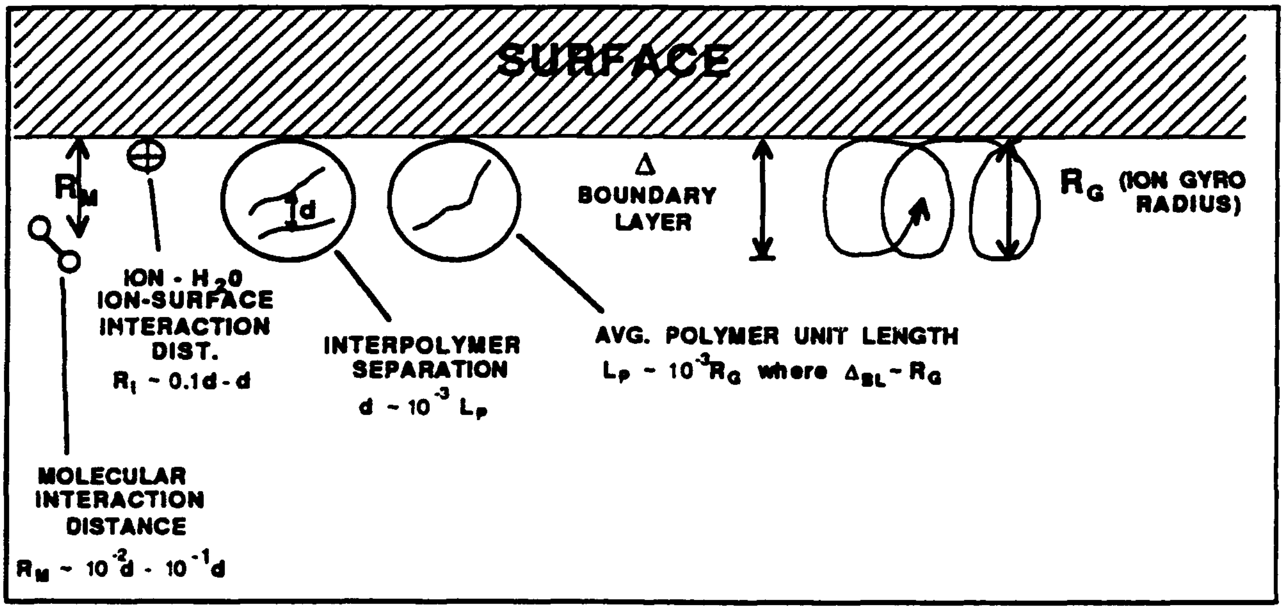

Figure 1. Comparative Scale of Meaningful Distances for Polymers in Salt Water

The presence of ions along the polymer main chain (or carbon "backbone") can provide a mechanism for "gettering" or "scavenging" strongly interacting species from the first surface layer, and thus weakening the attraction of the surface for the nearby polar water molecules. The Team also believes that the presence of polar groups along the monomer unit of the polymer may enhance the ability of the dilute solution to absorb flow energy in that these off-chain groups serve as sinks for acoustic noise through pronounced interaction with the collective oscillations of the polymer because of the relatively strong electrostatic ion-dipole and dipole-dipole interactions. Clearly, fundamental explanations of these phenomena must be obtained if tailored polymers are to be "custom-made" for noise and drag reduction. This approach does suggest that the nature of the surface may be critical (i.e., bare metallic vs. painted metal vs. insulator) in determining the net frictional contribution of the first several H_2O layers.

Thus, the series of experiments performed by USBI (Ms. Murphree), although not meant to be comprehensive, are consistent with the ASTI Team's hypothesis regarding polymer efforts on surface/water interactions. The anionic form of the polymer provides a mechanism for removing the active species at the surface and weakening the interaction with the adjacent water layers in that region. The most effective drag-reducing polymer also possesses significant polar groups ($-NH_2$), but the relative results of the experiments suggest that this latter property may not be as important as the scavenging capability for active species, at least at these Reynolds numbers. The tailored selection of such monomer groups to optimize noise reduction by polymer application is clearly one of the most intriguing aspects of a polymer science program strategy.

It is also important to recognize that surfactants most probably assume a very different configuration (near spherical) in the dilute solutions and will most certainly affect a much more localized surface region than the chain polymer. A detailed comparison of the relative water/surface interaction effects of surfactants vs. polymer is of great interest, but beyond the scope of this report. Although it is attractive to consider the synergistic potential for using surfactants in combination with polymers (as suggested by Ms. Murphree of USBI), developing a semi-quantitative description of such interaction is extremely complex. The most reasonable next step is that a series of experiments should be planned to unravel the variations in behavior going from a simple polymer (e.g., PEO) through more complicated species (with polar "off carbon backbone" groups) in both salt and deionized water so as to provide some additional insights on energy transfer mechanisms.

INTRODUCTION

The focus of this project has been to investigate the mechanisms of drag reduction experimentally. We began with a literature survey to identify mechanistic theories. From our readings, we found that theories abound on the reasons that turbulence can be reduced by the introduction of small amounts of additives into the boundary layer, but agreement between any of them is rare. In the forty years since B.A. Toms first reported his observations of friction reduction due to additives, no definitive mechanism has been accepted for the phenomenon. This fact may suggest that not one but several mechanisms are responsible, perhaps working synergistically to reduce turbulence.

In the case of polymeric drag reduction, most researchers generally agree that the mechanism involves macromolecular extension; in the case of surfactants, fibers and other materials, a similar process may take place. The role these extended structures may play in the reduction of drag is unknown. It may involve purely mechanical results of their presence in the boundary layer; the presence of additive molecules may alter normal mass transport toward or away from the substrate surface; additives may absorb energy which might otherwise create turbulence*; they may affect the orientation and alignment of the water molecules in the boundary layer**; or they may adsorb to the surface, changing its physical properties.

The first we call the "energy absorption" hypothesis. This is the idea that drag-reducing additive molecules in the boundary layer absorb energy from the bulk solvent. Transition to turbulence occurs when the rate at which energy enters the boundary layer exceeds the rate at which the additive molecules can absorb that energy. Energy might be absorbed mechanically, through chain extension, aggregate expansion, molecular strain and other means, or chemically, through temporary higher energy bonds, intermolecular associations, etc.

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* Energy absorption may take place as a result of a variety of mechanisms and is not exclusive of mechanical action or alignment of the H_2O molecules.

** This is the Team's preferred hypothesis since it can be approached at the molecular interaction level.

Another potential mechanism is the "hydrophilic area" hypothesis. An "adsorbed-entangled layer" has been shown to exist in which aggregates of drag-reducing material are temporarily adsorbed onto the flow boundary. These adsorbed aggregates may affect the structure of the surrounding fluid, interact with eddies, or play some other role in drag reduction.

The third mechanism is called the "water structure" hypothesis. It has been shown that if a substrate has charged or partially charged sites along its surface, polar solvents, like water, will align themselves accordingly. If the charges are properly spaced and the solvent is quite polar, this orderly alignment can extend through many molecular layers of solvent. If water structure in the boundary layer does indeed play a role in drag reduction, then polymeric coatings with slight anionic or cationic charges should show friction reduction and neutral coating binders should show no effect at all. Another means of structuring the water at the surface would be to suspend long-chain polymer branches from the backbone of a coating binder. The properties of these side-branch polymers would certainly have an effect on the water molecules surrounding them. They could be designed to have varying degrees of hydrophobic or hydrophilic character or to have ionic charges spaced along their length. Though linear polymers are generally better drag-reducers than branched ones in additive studies, pendant chains might work well as a coating on a substrate.

Using these ideas as guides, several experiments were run using the rotating disk drag tester at USBI in Huntsville, AL. A selection of polymers and surfactants were tested by adding them to a cylindrical tank of water and measuring the torque required to rotate a disk in the tank. Using torque and temperature measurements, Reynolds number and friction factor (the ratio of torque to the product of disk radius, disk surface area and stagnation enthalpy of flow, in units of L^3) were calculated and graphed (see Figures 6-13).

EXPERIMENTAL

The following materials were tested to investigate their effect on drag:

- Polyacrylamide

Nonionic polyacrylamide, molecular weight = 5-6 million

- Praestol 2540

Stockhausen proprietary anionic acrylamide copolymer; solution viscosity at 0.5% in distilled water is greater than 5500 cP.

- Oraestol 2530

Stockhausen proprietary anionic acrylamide copolymer; solution viscosity at 0.5% in distilled water is greater than 4500 cP.

- Algaeton

Exopolysaccharide derived from red algae. Obtained from Princeton Polymers, Inc.

- Polyox

Polyethylene oxide product of the Union Carbide Corporation.

- Fluorad surfactants

A family of fluorine-containing additives for coatings and other systems; selected to investigate whether the fluorine might add some drag-reducing properties.

FC430	nonionic liquid surfactant; compatible with aqueous and nonaqueous systems; viscosity approximately 15000 cP using #3 spindle (Brookfield) at 6 rpm.
FC431	nonionic liquid surfactant in ethyl acetate; compatible with polar organic solvents; Brookfield viscosity ≈ 200 cP using #1 spindle at 60 rpm.
FC120	anionic fluorosurfactant supplied in a 25% active mixture of ammonium perfluoralkylsulfonates in a 50/50 blend of water and ethylene glycol monobutyl ether; compatible with aqueous systems; viscosity ≈ water.
FC100	amphoteric surfactant supplied in a 25% solution in a water/diethylene glycol monobutyl ether mix; compatible with aqueous systems; viscosity ≈ water.
FC740	nonionic fluoroaliphatic polymeric esters supplied in a 50% solution in an unspecified solvent, possibly aromatic naphtha; compatible with low polarity organic solvents; low viscosity.
135	fluorochemical alkyl quaternary ammonium iodides in a 50% solution with water and isopropyl alcohol; compatible with aqueous systems; low viscosity.
170-C	nonionic liquid surfactant composed of a mixture of fluorinated and nonfluorinated polyethylene ethanols; compatible in polar solvents including water.

- Poly(acrylamide-co-diacetone acrylamide) (p-AM-co-DAAM)

Produced by project personnel according to the methods of C.L. McCormick[1]. In the referenced paper, this material is shown to exhibit significant drag reduction,

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[1] McCormick, C.; Hester, R.; Morgan, S.; and Safieddine, A. Macromolecules 23, 8 (1990): 2124-2131.

particularly in the presence of electrolytes. The 35% DAAM/65% AM variation was synthesized.

In most cases, the materials were tested as received from the manufacturer. The polyacrylamide was recrystallized from methanol and all reactants for the (p-AM-co-DAAM) were recrystallized from either methanol or water, as appropriate. All materials were dissolved or suspended in water before being added to the 15.925 liter tank to make the appropriate concentration of solution.

The rotating disk test device consisted of a motor driving a shaft on the end of which was suspended a smooth 9-inch disk. The same disk was used for all tests. The shaft and disk were lowered into a tank of water and rotated with measurements of torque and temperature being taken at intervals of 200 rpm within a range of 500 to 1500 rpm. Two runs were made of each experiment and the following calculations[2] were made from the observations:

$$\text{Reynolds number} = \frac{R^2\Omega\rho}{\mu}$$

where
R = radius of the disk
Ω = angular velocity
ρ = density of the fluid (corrected for temperature)
μ = viscosity of the fluid (corrected for temperature)

$$\text{Coefficient of friction Cf} = \frac{T}{RAK}$$

where
T = torque required to rotate the disk
A = surface area of the disk (both sides)
K = $\frac{\rho R^2\Omega^2}{2} = \frac{\rho u^2}{2}$

Photos of the improved version of the experimental apparatus are included in Figures 2 and 3. Schematics of the original and improved version of the apparatus are also included, in Figures 4 and 5.

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[2] Byrd, R.B.; Stewart, W.E.; Lightfoot, E.N. <u>Transport Phenomena</u>. New York: Wiley, 1960, p. 203.

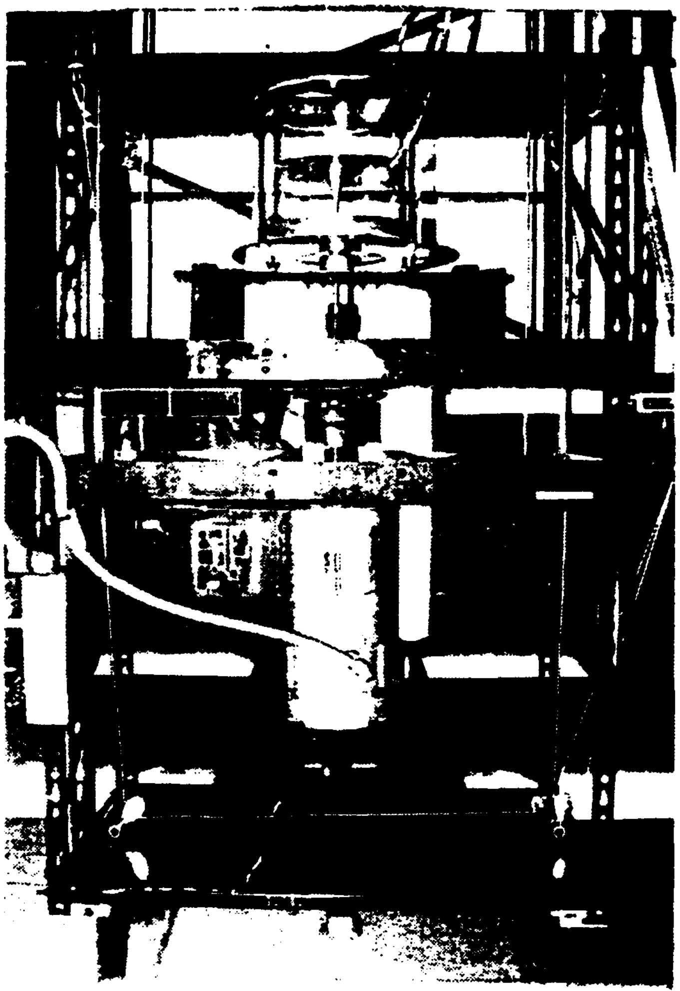


Figure 2. Experimental Apparatus

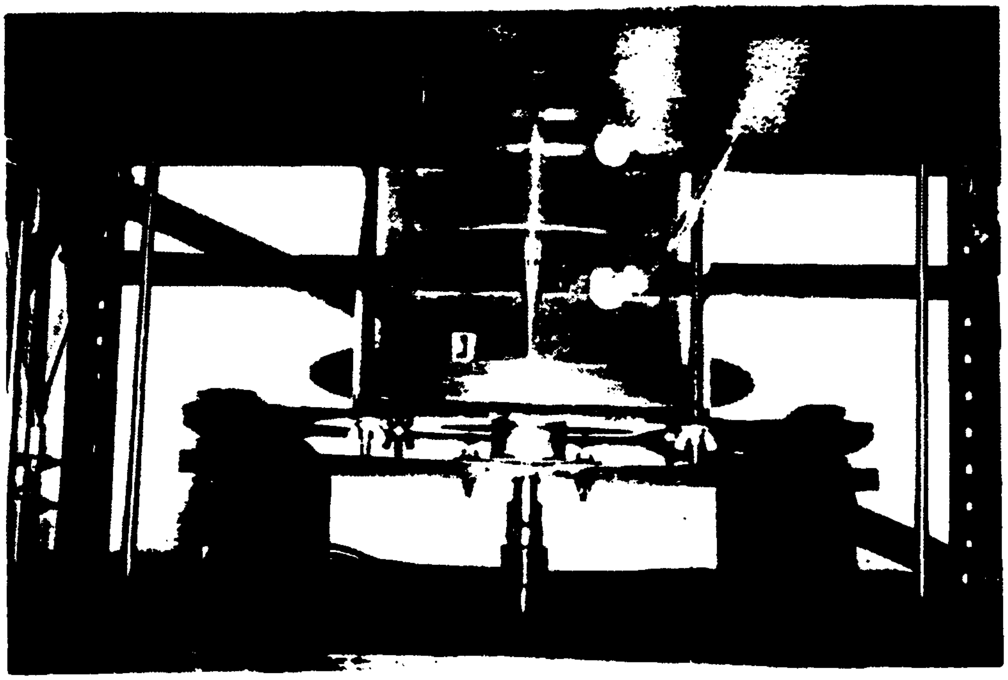


Figure 3. Experimental Apparatus Detail

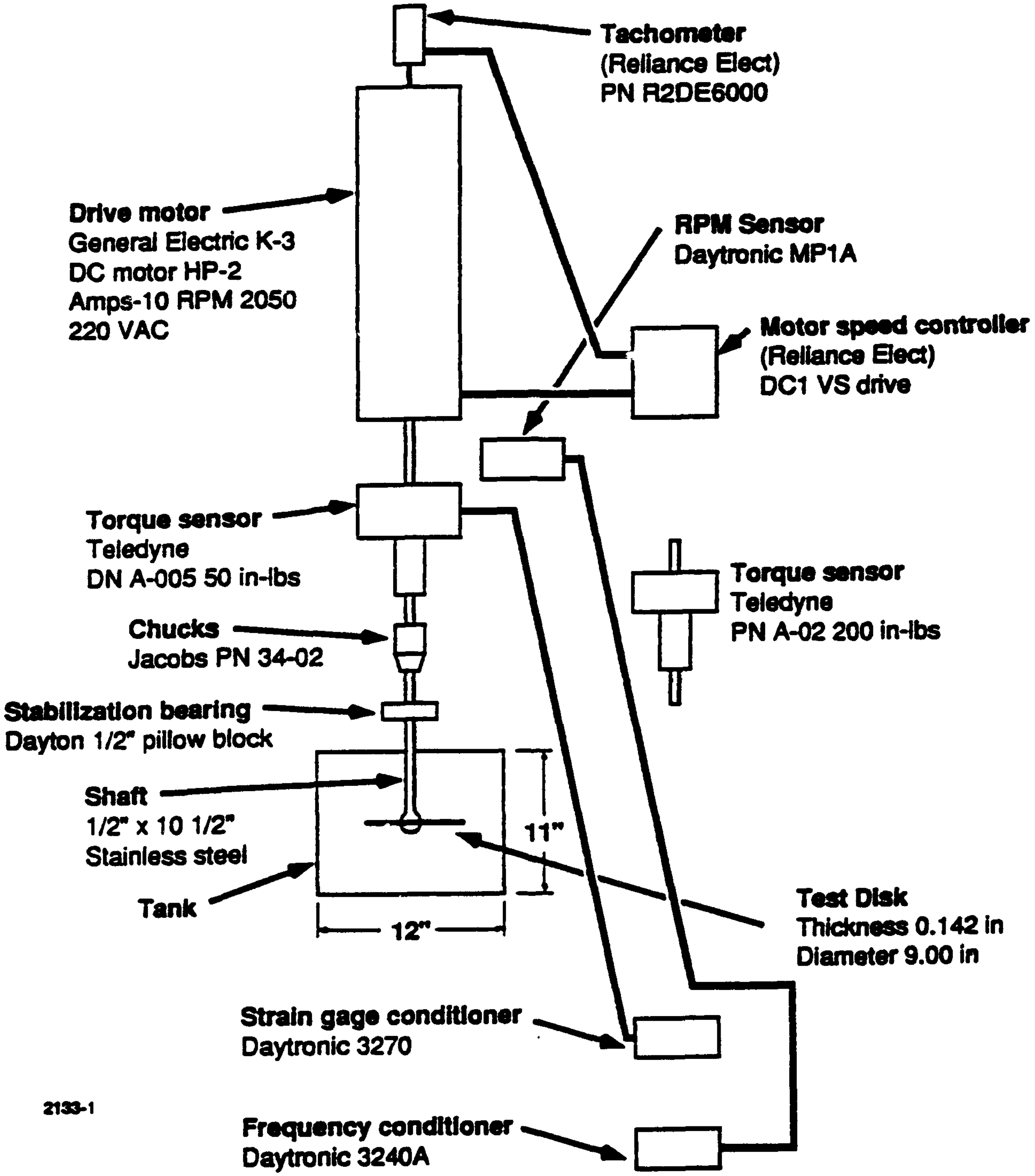
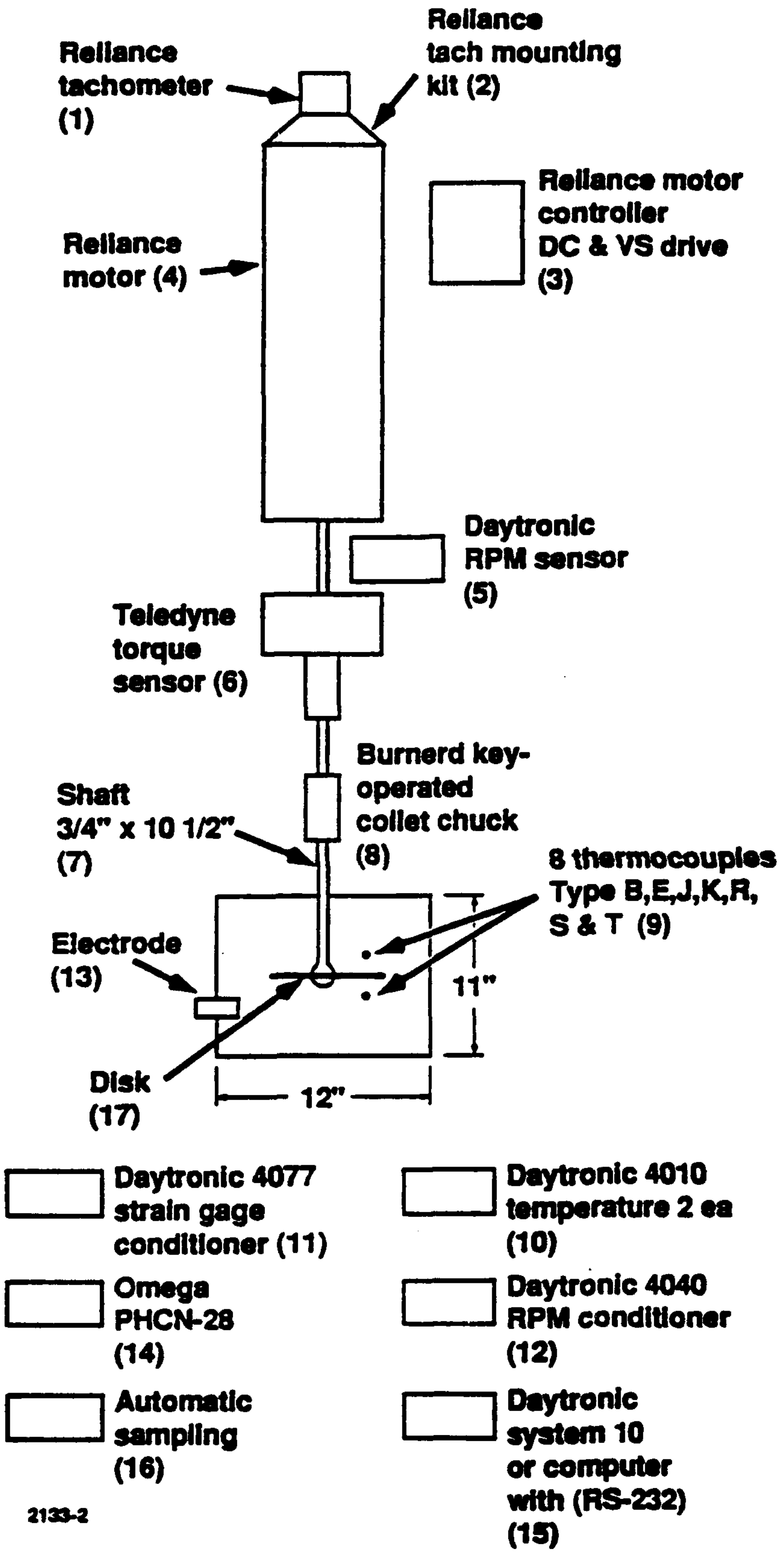


Figure 4. Drag Tester



1. Reliance tachometer R20. On hand.
2. Reliance tachometer mounting kit 417708-10 (for installation of tach. to motor). New purchase.
3. Reliance motor controller DC & VS drive on hand. R232 controllable not available at this time.
4. Reliance motor TI8R1117 2HP manufactured for tachometer installation (upright operation). New purchase.
5. Daytronic RMP sensor MP1A magnetic pick-up on hand.
6. Teledyne torque sensor A-005 50 in-lbs on hand.
7. Shaft 3/4" x 10 1/2" stainless steel constructed from single bar stock on one center. New purchase.
8. Burnerd key-operated collet chuck. 1" max cap type ea. Collet type ea 3/4".
9. Thermocouple Omega.
10. Daytronic 2 ea 4010 temperature meters. Units are standalone or operated on a R232 bus.
11. Daytronic 4077 strain gage conditioner. Standalone or R232.
12. Daytronic 4040 RPM conditioner. Standalone or R232.
13. pH electrode Omega used with PHCN-28.
14. pH meter PHCN-28 Omega. Standalone or R232.
15. Daytronic system 10 controller R232 or computer of like kind.
16. Automatic sampling. No information.
17. Test disk thickness 0.142 in. Diameter 9.00 in.

Drag tester mounting rack will be designed after all items are available.

Figure 5. Improved Drag Tester

RESULTS AND CONCLUSIONS

The following experiments were conducted. Two runs were made of each and the final values averaged. The drag reduction measurements in these experiments are plotted versus Reynolds numbers in Figures 6 through 13.

Experiment no. 1
Polymer: 5.00 ppm Polyacrylamide in D.I. Water

- as a baseline value for p-AM; upward slope at higher Reynolds numbers is due to equipment malfunction. All of the nonionic polyacrylamide experiments resulted in some drag reduction, though they were not the best drag-reducers tested.

Experiment No. 2
Polymer: 10.00 ppm Polyacrylamide in D.I. Water

- after Experiment No. 1, additional p-AM was added to the tank to make a 10 ppm solution. Drag increase from 5 ppm solution in Experiment No. 1 could be caused by shear degradation of the polymer from the first experiment.

Experiment No. 3
Polymer: 5.00 ppm Polyacrylamide in Instant Ocean (synthetic seawater)

- to investigate the effect of electrolytes on p-AM; crag increased from that exhibited in D.I. water.

Experiment No. 4
Polymer: 10.00 ppm Polyacrylamide in Instant Ocean

- after Experiment No. 3, additional p-AM was added to the tank to make a 10 ppm solution; drag increased as in Experiment No. 2.

Experiment No. 5
Polymer: 5.00 ppm Praestol 2540 in D.I. Water

- An Ionic polyacrylamide, this solution showed the best drag reduction of all materials tested.

Experiment No. 6
Polymer: 5.00 ppm Praestol 2530 in D.I. Water

- This material also showed excellent drag reduction, though no as much as the 2540.

Experiment No. 7
Polymer: 5.00 ppm Praestol 2530 in D.I. Water

- This is the same materials as No. 6 above, except that the polymer had been dissolved in water for 15 days before test. This may have served to better solvate the polymer because the drag reduction here is slightly better than for Experiment No. 6.

Experiment No. 8
Polymer: 4.98 ppm Algaeton in D.I. Water

- The only natural material in this study, it showed only slight drag reduction. One reason for this may be that the manufacturer (Princeton Polymer Laboratory) did not have the capability of fractionating the polymer and the material used was the natural exopolymer, probably containing a variety of molecular weights.

Experiment No. 9
Polymer: 4.98 ppm Algaeton in D.I. Water

- This experiment was run with the same tank of material as No. 8 in an effort to see the effects of any possible shear degradation. The drag reduction was less than in No. 8.

Experiment No. 10
Polymer: 5.04 ppm Praestol 2530 in D.I. Water

- Here a fresh (not aged) solution of 2530 was used to No. 6 and No. 7 above after a realignment of the equipment. The drag reduction was somewhat better than No. 7.

Experiment No. 11
Polymer: 5.18 ppm Polyox in D.I. Water

- A fresh solution of Polyox was tested and, as expected, it was one of the best drag-reducers. Note, however, that the Praestol 2540 showed even lower friction.

Experiment No. 12
Surfactant: 5.13 ppm Fluorad FC430 in D.I. Water (nonionic)

- Although this was one of the better drag-reducers among the surfactants, all the surfactants either showed negligible drag reduction or indeed an increase in drag. None of them had a great effect either way. It is possible that they might perform better in conjunction with surfactants.

Experiment No. 13
Surfactant: 5.13 ppm Fluorad FC430 in D.I. Water (nonionic)

- The same tank of solution from No. 12 was tested again to check for degradation. As expected, since surfactants generally don't exhibit shear degradation, the performance was almost identical to No. 12.

Experiment No. 14
Surfactant: 4.98 ppm Fluorad FC431 in D.I. Water (nonionic)

- The performance of this material was not remarkable.

Experiment No. 15
Surfactant: 10.42 ppm Fluorad FC431 in D.I. Water (nonionic)

- A fresh 10 ppm solution was tested. Its performance was similar to that of the 5 ppm solution (No. 14).

Experiment No. 16
Surfactant: 10.42 ppm Fluorad FC431 in D.I. Water (nonionic)

- The same tank of solution was retested to check degradation effects; drag did increase slightly but not significantly over that in No. 15.

Experiment No. 17
Surfactant: 5.11 ppm Fluorad FC120 in D.I. Water (anionic)

- The performance of this material was not remarkable.

Experiment No. 18
Surfactant: 5.11 ppm Fluorad FC100 in D.I. Water (amphoteric)

- The performance of this material was not remarkable.

Experiment No. 19
Surfactant: 5.41 ppm Fluorad FC100 in D.I. Water (amphoteric)

- A fresh solution was tested; performance was similar to No. 18.

Experiment No. 20
Surfactant: 5.22 ppm Fluorad FC740 in D.I. Water (nonionic esters)

- The performance of this material was not remarkable.

Experiment No. 21
Surfactant: 5.02 ppm Fluorad 135 in D.I. Water (quaternary ammonium salt)

- The performance of this material was not remarkable.

Experiment No. 22
Surfactant: 5.01 ppm Fluorad 170-C in D.I. Water (polyethylene ethanols)

- The performance of this material was not remarkable.

Experiment No. 23
Polymer: 10.35 ppm Polyox in D.I. Water

- This is a test of a 20-day-old solution of Polyox. The 10 ppm aged solution performed almost identically to the 5 ppm fresh solution. Although a higher concentration was necessary to achieve the same reduction in friction, this is contrary to some studies which have shown almost no drag reduction for Polyox solutions only a day or so old.

Experiment No. 24
Polymer: 6.05 ppm Poly(acrylamide-co-diacetone acrylamide) in D.I. Water

- This material showed some drag reduction, but was not one of the best. One reason for this could be that, like the Algaeton (nos. 8, 9) it was tested unfractionated. Better drag reduction (such as McCormick found) might well be shown by the higher molecular weight fractions.

Experiment No. 25
Polymer: 6.20 ppm Poly(acrylamide-co-diacetone acrylamide) in D.I. Water

- A fresh solution showing better drag reduction than No. 24. Higher concentrations could well show even better results.

Experiment No. 26
Polymer: 6.20 ppm Poly(acrylamide-co-diacetone acrylamide) in D.I. Water

- This is the same tank of solution as in No. 25, tested again to check for shear degradation. This run showed much less drag reduction than seen in the first run (No. 25).

CONCLUSIONS

In general, polymers performed better than surfactants. The best materials tested were the ionic Stockhausen's Praestol 2540 and 2530 and Union Carbide's Polyox. Nonionic polyacrylamide gave fair drag reduction; its copolymer with diacetone acrylamide showed some capability, but would probably give better results if only the higher molecular weight fractions were used. The exopolysaccharide Algaeton gave some reduction in drag but, likewise, would probably be more effective using the higher molecular weight fractions. None of the fluorosurfactants tested exhibited any capability whatsoever, although they might prove more effective if combined with polymers.

The result of an ionic polyacrylamide performing better than a non-ionic polyacrylamide in the previous experiments raises the question of the ion's role in drag reduction. If, in fact, ionic species do enhance the reduction of drag in a system, the mechanism by which this occurs is not clear. Further experimentation and investigation is necessary to characterize the mechanism. In a more restricted sense, the results may also provide some information on the limited role of additional degrees of internal molecular freedom on enhancing drag reduction at high Reynolds numbers.

The ion effects on the "robustness" of the molecular structure (i.e., its capability to repetitively provide drag reduction) have been investigated for certain polymers by Dr. Kim at the Naval Research Laboratory. Ultimately, it will be necessary to obtain similar data on other polymers, such as those studied in the USBI experiments, so as to identify and illuminate potential operational constraints (e.g., project performance in salt water). The CORTANA Team's view is that, although deionized water experiments are critical to isolating ionic effects in determining robustness, the major operational screening test for the polymer should be in salt water.

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1. Byrd, R.B.; Stewart, W.E.; and Lightfoot, E.N. Transport Phenomena. New York: Wiley, 1960.

2. McCormick, C.; Hester, R.; Morgan, S.; and Safieddine, A. Macromolecules 23, 8 (1990): 2124-2131.

3. Morgan, S., and McCormick, C.L. "Water-Soluble Copolymers XXXII: Macromolecular Drag Reduction. A Review of Predictive Theories and the Effects of Polymer Structure. Progress in Polymer Science 15 (1990): 507-549.

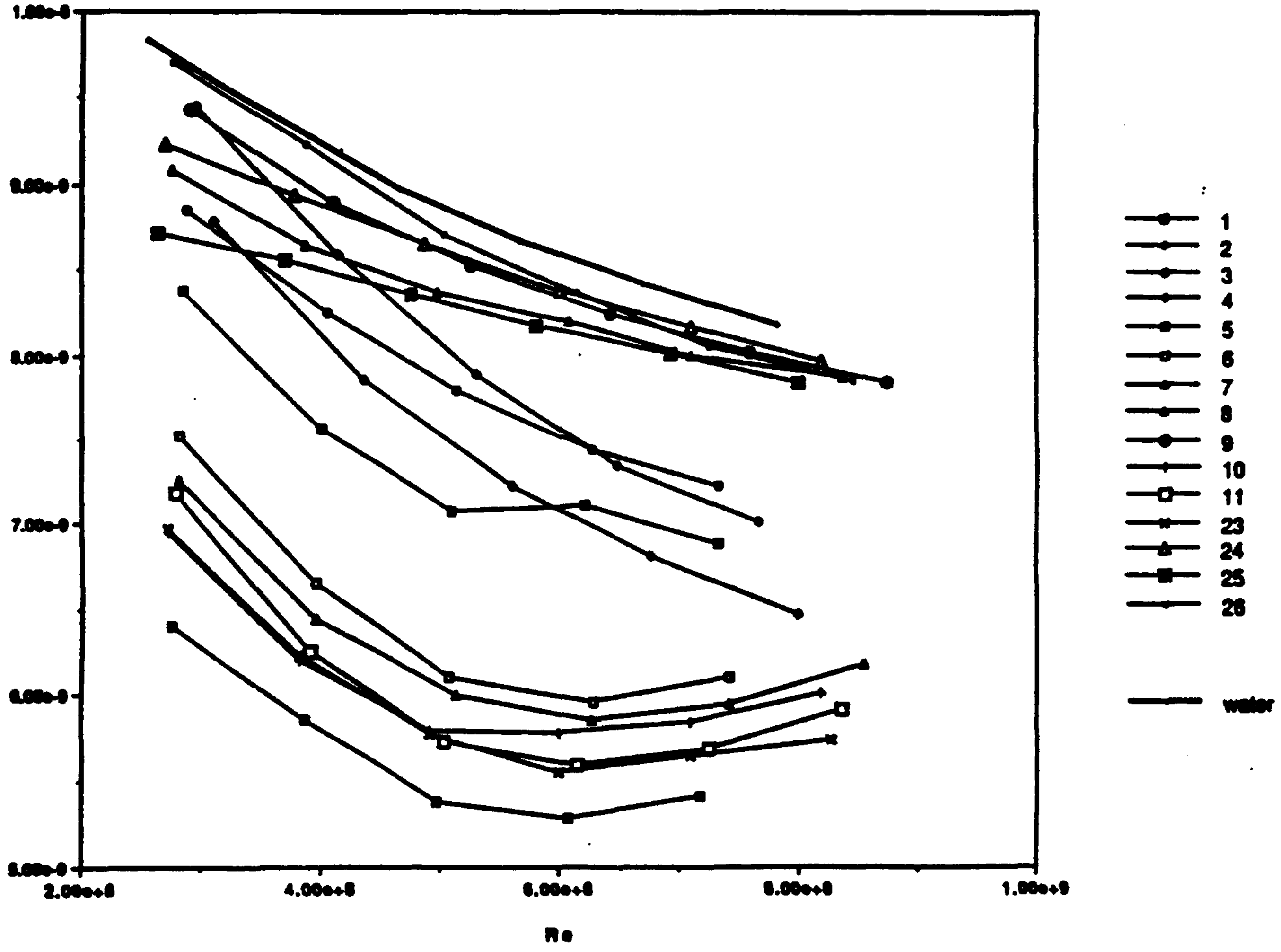

Figure 6. Additive Drag Data: Polymers

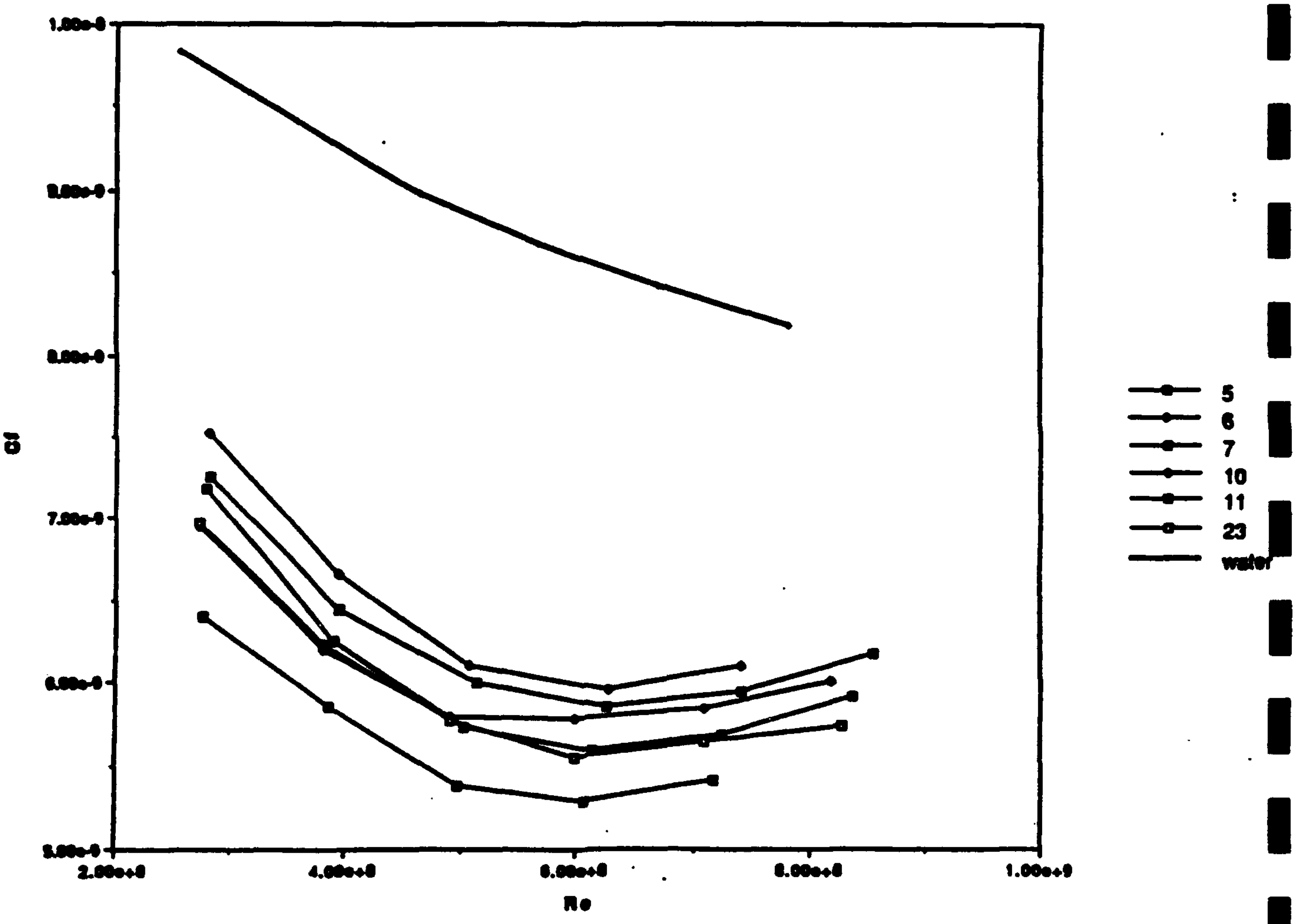

Figure 7. Most Effective Additives

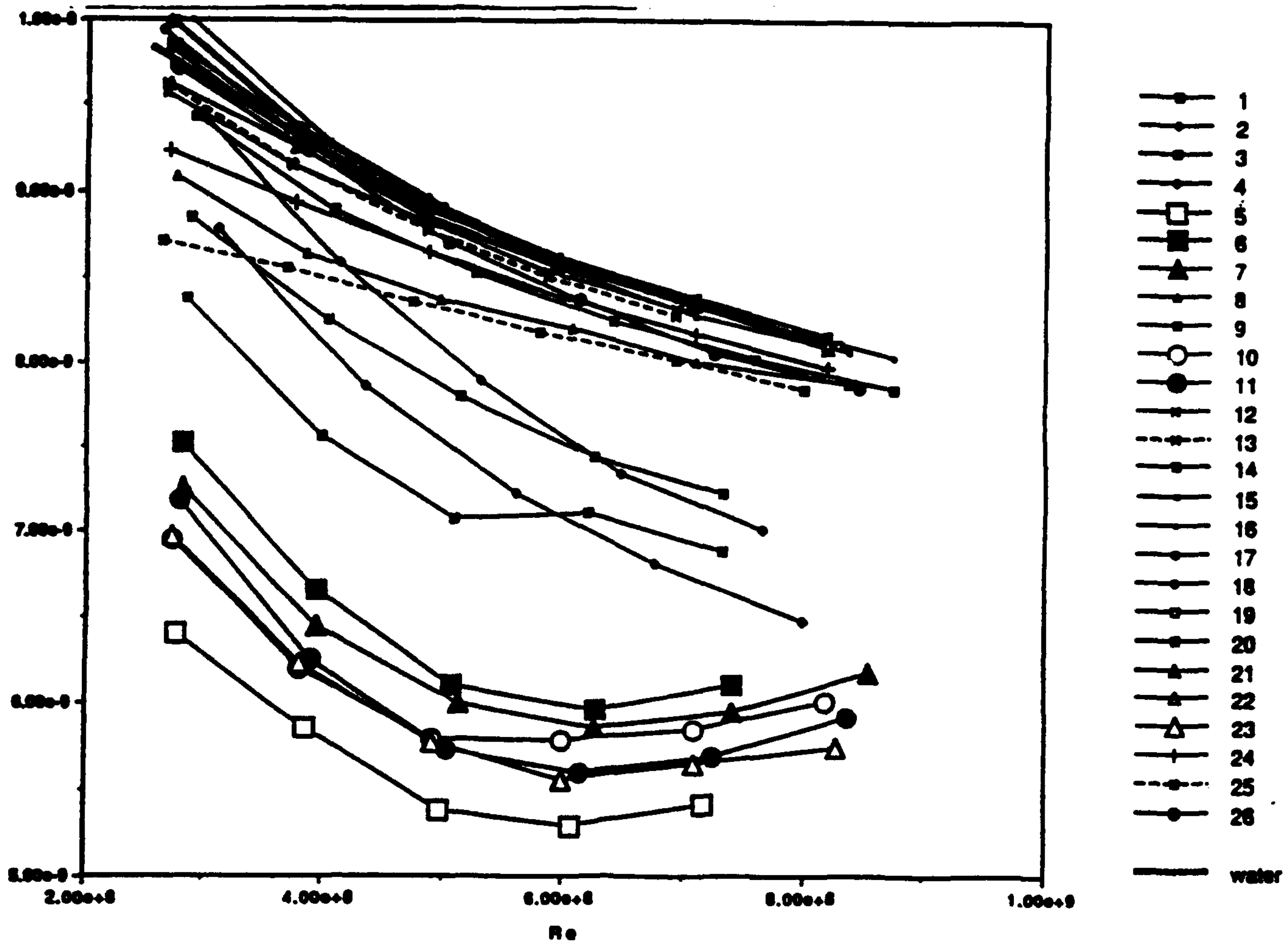

Figure 8. Additive Drag Reduction Experiments

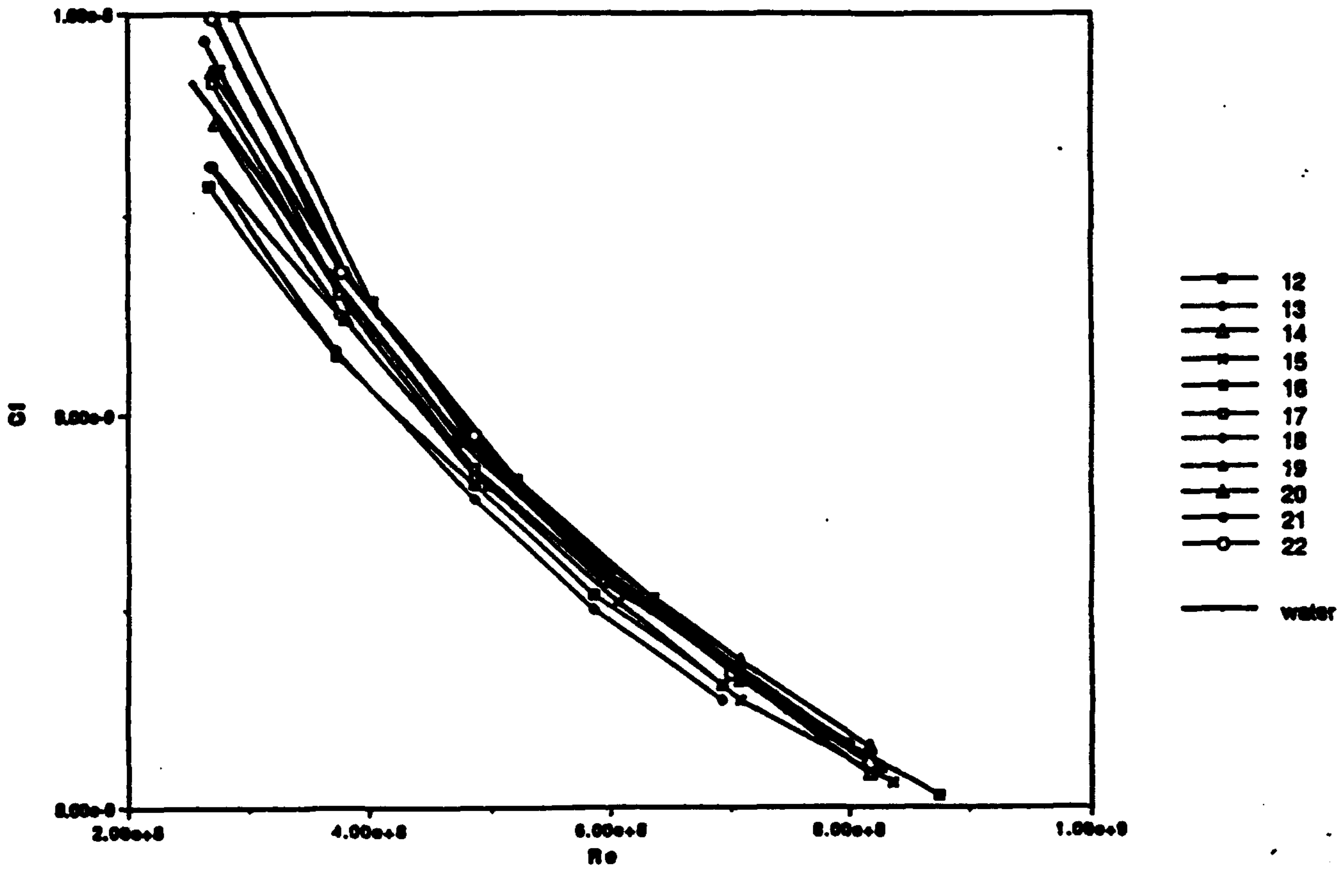

Figure 9. Additive Drag Data: Surfactants

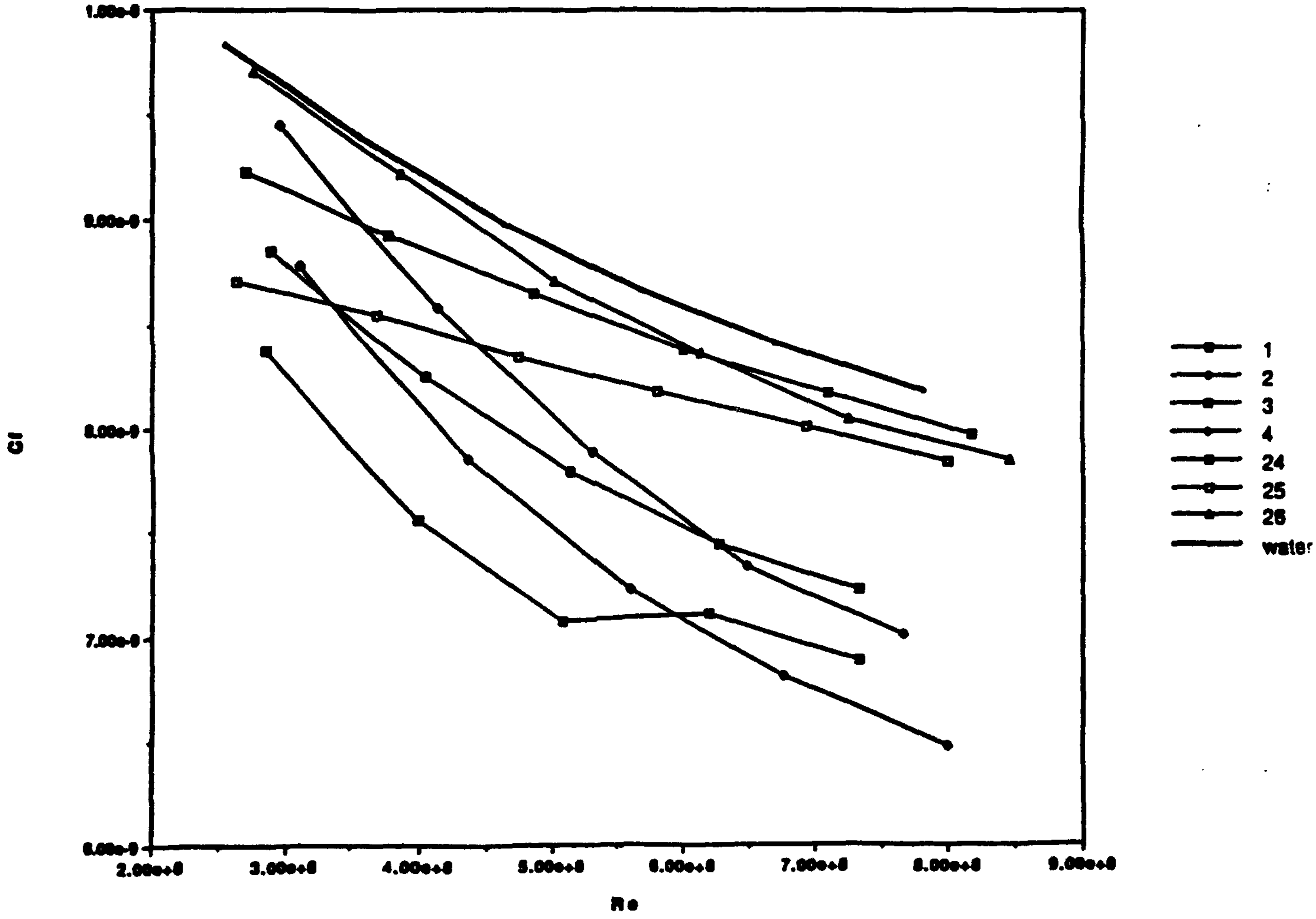

Figure 10. Polyacrylamide vs. Poly(acrylamide-co-diacetoneacrylamide)

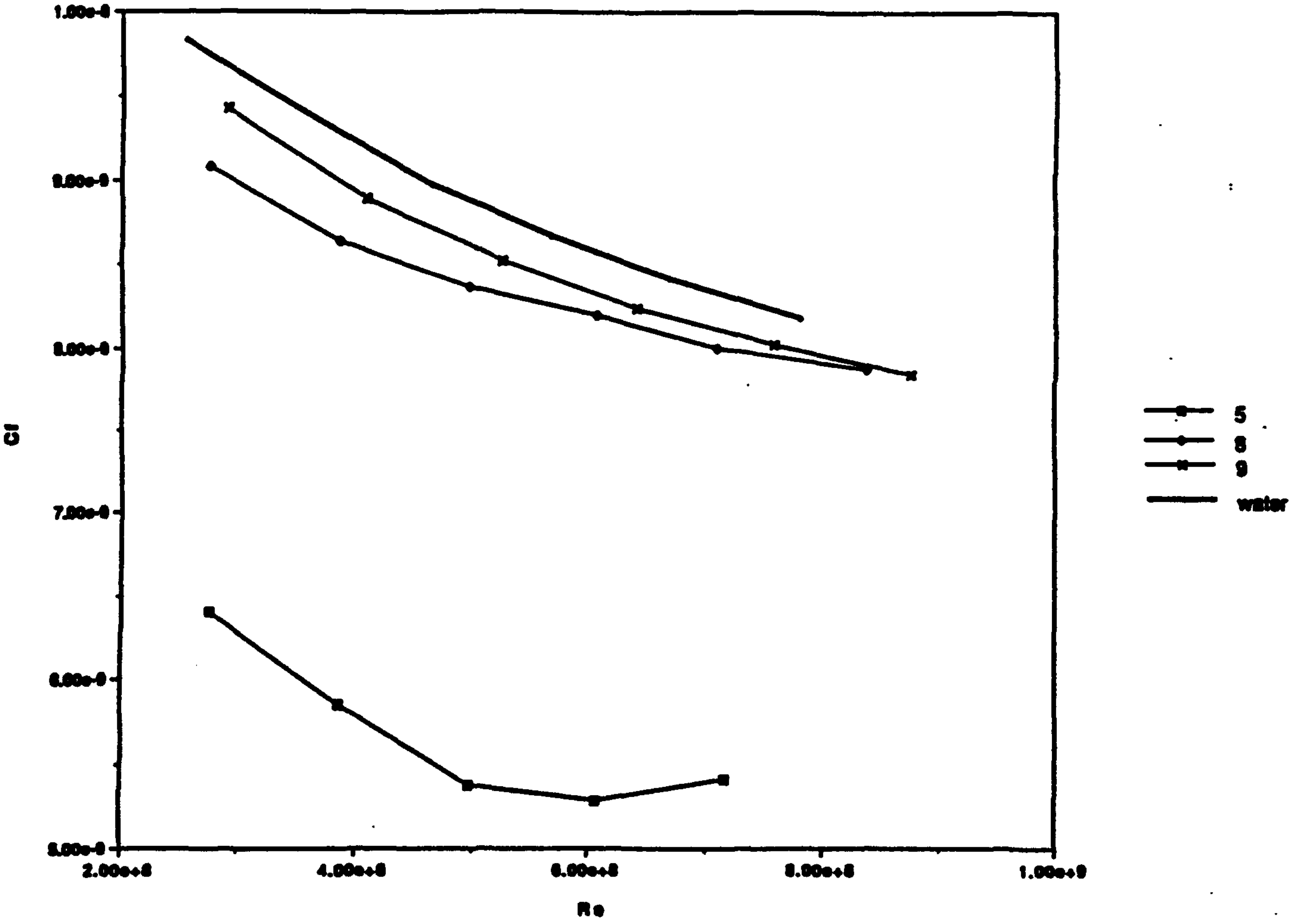

Figure 11. Natural Material Compared to Best Synthetic

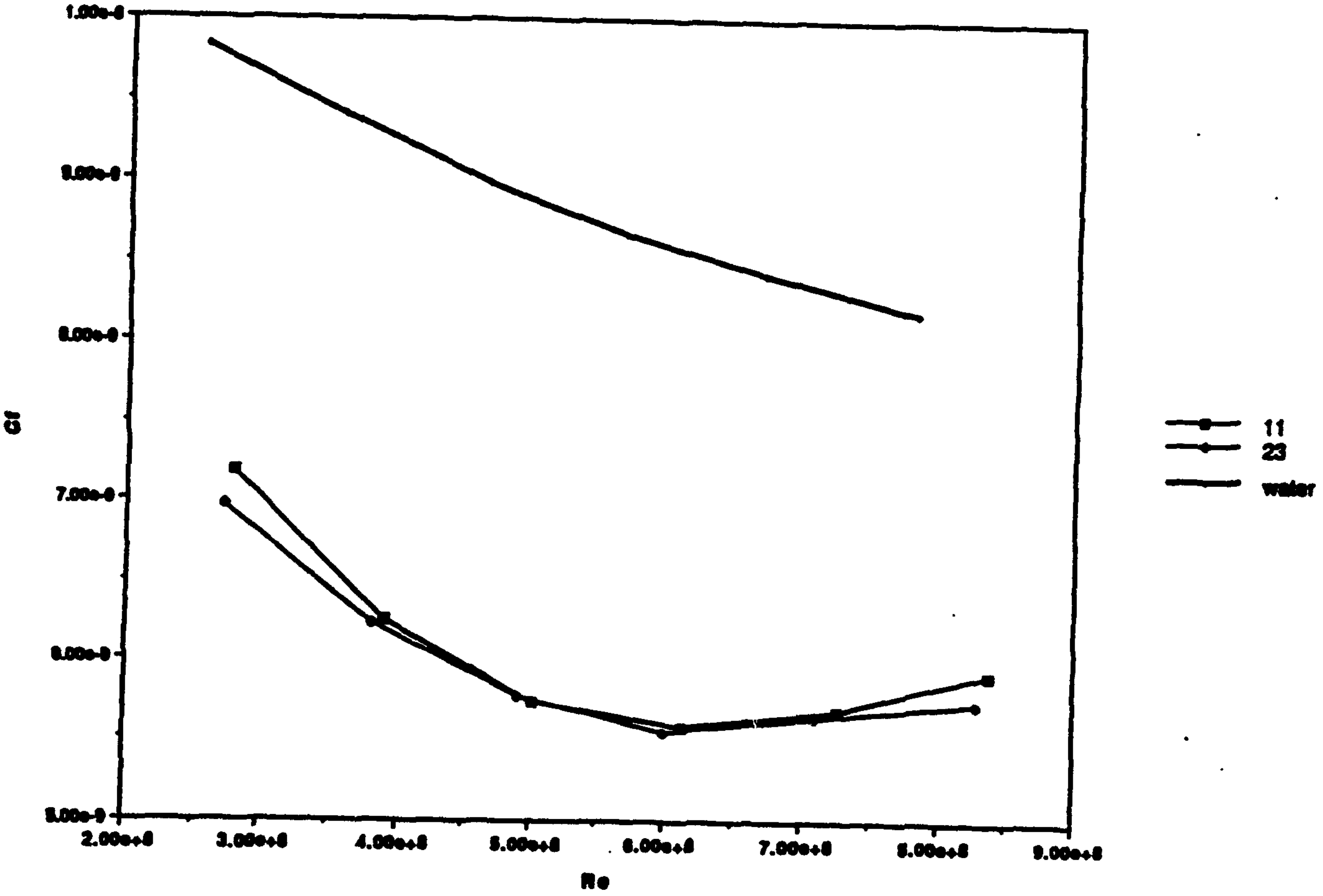

Figure 12. Polyox: Fresh vs. Aged

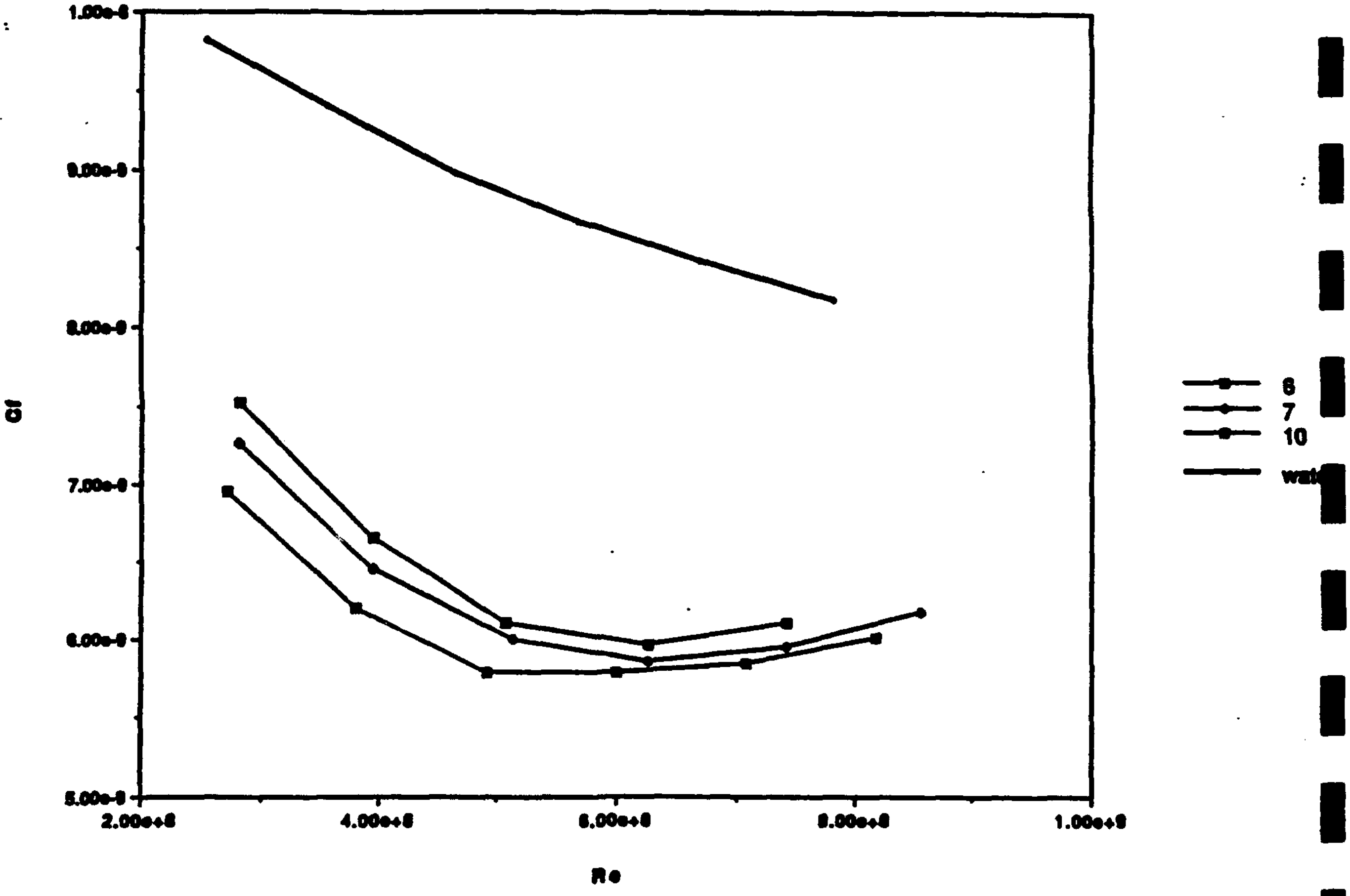

Figure 13. Praestol 2530: Fresh vs. Aged