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PN-ACM-339

# Final Report

## August 16, 1991 – December 31, 1999

## I. Executive Summary

This collaborative research project had a number of major objectives which were met:

1. In order to utilize surfactant drag reducing additives in district heating systems, the relationships among cationic surfactant chemical structures, counterion chemical structures, their concentrations and their molar ratios on micellar size and shape (microstructure), rheology and drag reducing effectiveness (percent reduction and effective temperature range) must be known. Experimental studies at the Czech Academy of Sciences and at Ohio State University have clarified the relationships. We have also examined non-ionic and zwitterionic/anionic surfactants. Nine papers on the results have been published or accepted in archival journals, fourteen in Proceedings of International Conferences and twenty paper presentations have been made at national and international conferences.
2. As part of the studies of drag reduction mechanisms and drag reduction effectiveness, two surprising results were obtained. We recognized that data on some of our systems lay well below (higher drag reduction) the maximum drag reduction asymptote proposed by Virk for high polymers nearly 30 years ago[1]. The new asymptote extended maximum drag reduction limits to over 90%. Mean turbulent velocity profiles with limiting slopes twice that proposed by Virk[1] for high polymers were discovered. These remarkable results, as well as differences in details of statistical turbulence phenomena, imply differences in the mechanisms of drag reduction by surfactants vs. high polymers, a concept that we were the first to propose.

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3. Heat transfer data were obtained on a cationic surfactant system and a zwitterionic/anionic system. A novel heat exchanger tube design (fluted tubes) which minimizes the adverse heat transfer reduction effect of drag reducting surfactant systems was demonstrated. The greater effectiveness of plate heat exchangers vs. tube-in-tube heat exchangers was also demonstrated.
4. A successful field test of Hoechst's Habon G cationic surfactant additive was carried out in the secondary system of a 250-flat district heating system in Kladno-Krocehlavy (near Prague) during the winter of 1993-94. Reduction in energy losses of about one third were obtained. Unfortunately, Hoechst was not able to obtain environmental approval of this additive in Denmark and Germany because of its relatively slow rate of biodegradation and they abandoned further development work on it. Recently, Akzo Nobel developed a zwitterionic/anionic surfactant product wich biodegrades rapidly. It has recently been approved for use in district heating systems in Denmark. A field test of this additive in a building at the Institute of Hydrodynamics of the Czech Academy of Sciences is planned for winter 2000-2001 and we will continue to collaborate on the field test.
5. Improved infrastructure, particularly computers, computer software (FLUENT), turbulence measurement equipment, and rheological equipment for the Czech investigators have come from funds from this project. Three junior Czech scientists spent 3-4 months each at Ohio State working on the project. The co-PI's Dr. Myska (2 weeks) and Professor Pollert (one week) also worked on the project at Ohio State. They and other Czech colleagues gained exposure by attending international meetings in the U.S.(2), Italy(1), Spain(1), United Kingdom(1), Slovenia(1), Israel(1) and Czech Republic(2) to present papers on research completed under this project. A major conference on Drag Reduction was held in Prague in September 13-15, 1999. Professor Pollert chaired the conference and Dr. Chara, a collaborator from the Institute of Hydrodynamics, was Organizing Chair. The senior Czech collaborators from both the Czech Technical University and from the Institute of

Hydrodynamics (Professor Pollert , Dr. Myska, and Dr. Chara) are now recognized internationally for their research on surfactant drag reduction.

## II. Research Objectives

The goals of the project were to study the effects of surfactant chemical structure, counterion chemical structure, their concentrations and ratios on drag reduction effectiveness, to investigate the mechanism of surfactant drag reduction, to develop techniques to reduce the adverse effect drag reducing systems have on heat transfer, to demonstrate by a field test that drag reducing surfactants could successfully reduce energy requirements in district heating systems and to improve the infrastructure, experimental, and computational capabilites of Czech scientists at the Czech Technical University and the Institute of Hydrodynamics of the Czech Academy of Sciences, to assist senior scientists in taking active roles in the international research community and to train junior scientists in modern research techniques by several month studies at Ohio State. The project was supported by grants from the Czech government to both the Czech Technical University and the Institute of Hydrodynamics, and from the International Energy Association, the U.S. Department of Energy, the American Chemical Society-Petroleum Research Fund, and The Ohio State University to Prof. Zakin.

A list of the Proceedings and archival journal publications resulting from this research is given in Section V. Each publication contains relevant pertinent literature references and reprints are attached.

## III. Methods and Results

Measurements of the following quantities were made in laboratory experiments:

1. Percent reductions in turbulent friction factors as a function of solvent Reynolds numbers over a range of temperatures and in tubes of different diameters for different surfactant solutions. These meaurements were made in tube test sections in recirculation systems.
2. Percent reductions in heat transfer coefficients in tube-tube and plate heat exchangers for several different surfactant solutions. These measurements were made in recirculation systems.
3. Turbulence and velocity profile measurements were made in a 40 mm diameter flow loop using Dantec LDV equipment.
4. Rheology behaviors of different surfactant solutions were measured over ranges of temperatures. These included shear velocity, first normal stress differences ($N_1$), dynamic viscosities, apparent extensional viscosities, and flow bisefringence. These measurements were made in Couette and cone-and-plate rheometers and in an opposing nozzle extensional viscometer and in a Couette flow birefringence cell.
5. Micellar sizes were obtained by dynamic light scattering and microstructures by cryogenic Transmission Electron Microscope (cryo-TEM) imaging.
6. A field test in the secondary system of a small district heating system with about 250 flats measured the reduction in energy requirements, the make-up of surfactant required, and the effective life of the surfactant additive.

Major Scientific Findings:

1. The chemical structure of cationic surfactants and of their counterions, their concentrations and molar ratios affect turbulent drag reduction effectiveness, effective drag reduction temperature ranges, surfactant microstructure and rheological behavior. Effective

surfactant drag reducing solutions generally had microstructures made up of networks of elongated threadlike micelles.

2. Friction factors lower than the accepted Virk asymptote valid for high polymer drag reducing additives were observed. A new asymptote for surfactant systems was proposed. In addition, the limiting steepness of $u^+$ vs. $\ln y^+$ velocity profiles were found to be twice as large as the limiting profile for polymer solutions proposed by Virk[1].

3. Turbulence intensities of surfactant solutions in the axial direction were similar in magnitude but of different shape than those of water. Radial and tangential intensities were much lower than those of water.

4. Surfactant microstructures recovered after mechanical degradation in regions of high shear in times of the order of seconds. The recovery times depend on the surfactants' chemical natures, microstructures, concentrations, and the molar ratios of counterion to surfactant.

5. Heat transfer coefficients for surfactant drag reducing systems are reduced both in tube-tube and in plate heat exchangers, but by a much higher percentage in the former. This drawback to their use in district heating systems can be alleviated by use of fluted heat exchanger tubes.

6. A field test using a cationic surfactant (Habon G, manufactured by Hoechst AG) in the secondary system of a small district heating system of about 250 flats was successful. Energy requirements were reduced by a one third in this system and the solution, with make-up for leakage losses, was effective for at least one winter season after which the test had to be terminated. Unfortunately, Hoechst abandoned the product when environmental approval by Denmark was delayed because of its slow rate of biodegradation.

7. A new additive from Akzo Nobel, which is made up of a mixture of zwitterionic and anionic surfactants, and which biodegrades fairly rapidly, has recently been approved by Denmark. Our laboratory tests indicate that it is a promising additive and funds have been awarded by the Czech government for a field test at the Institute of Hydrodynamics next

winter. Design of the test conditions will be based on laboratory test results obtained at the Institute of Hydrodynamics and at Ohio State University. If this field test is successful, as expected, Akzo Nobel will collaborate in promoting the use of the drag reducing additive in commercial district heating systems in the Czech Republic and other eastern countries.

## IV. Impact

The technical impacts are described in Section III. The project has also had important impacts on engineers and scientists at the Institute of Hydrodynamics and at the Czech Technical University. The PI's, Professor Pollert and Dr. Myska, both spent time at Ohio State and have also been able to attend a number of international meetings at which they presented papers. Dr. Chara and Dr. Stern of the Institute of Hydrodynamics also presented papers at international meetings. Dr. Chara, P. Komrzy and J. Pollert Jr., each spent several months at Ohio State using modern laboratory equipment and computers and improving their English. Dr. Komrzy and Dr. Chara have completed their PhD's at the Czech Technical University and J. Pollert Jr. will complete his soon. (He was delayed as he had to serve in the Czech army for about one year.)

In addition, to broadening the scientific exposure of these personnel, a computer, computer software including FLUENT and laboratory equipment for drag reduction, turbulence and rheology experiments were provided to Professor Pollert, Dr. Myska, Dr. Chara, and Dr. Stern from this grant.

With their developing reputations in the field of drag reduction, the European Working Party on Drag Reduction, selected Professor Pollert to be Chair of the 11$^{th}$ Biennial Meeting held in Prague, September 13-15, 1999. Dr. Chara served as Vice-Chair and Program Director. The meeting was an excellent one with participants from European countries as well as Japan, Korea, and the U.S. I gave one of the Plenary Lectures with Dr. Myska as coauthor and described our important findings on the differences between polymer and surfactant drag reduction. Dr. Myska, Dr. Chara, and I each presented an additional paper (see Sector VII — Publications).

## V. Project Activities / Outputs

Presentations at international conferences and publications in Proceedings of these conferences and in archival journals are listed below. The meetings attended are noted in the Proceedings (also see summary in I). Training is described in IV.

**Presentations**

Z. Chara, J.L. Zakin, M. Severa, J. Myska "Turbulence Measurements of Drag Reducing Surfactant Systems" at 13th Biennial Turbulence Symposium, University of Missouri-Rolla, 1992.

J. Pollert, P. Komrzy, A. Vozenilek, J.L. Zakin, "Influence of Pipe Diameter and Temperature on Efficiency of Drag Reducing Surfactants," Academy of Sciences of the Czech Republic, Fluid Mechanics and Hydrodynamical Aspects of Biosphere, Castle Liblice, Sept. 20-21, 1993.

J. Myska, M. Severa, J. Zakin, "LDA Investigation of Turbulent Characteristics of Drag Reducing Surfactant Systems," Academy of Sciences of the Czech Republic, Fluid Mechanics and Hydrodynamical Aspects of Biosphere, Castle Liblice, Sept. 20-21, 1993.

J.L. Zakin, L.C. Chou, B.C. Smith, B. Lu, "Molecular Variables Affecting Drag Reduction by Cationic surfactants," Academy of Sciences of the Czech Republic, Fluid Mechanics and Hydrodynamical Aspects of Biosphere, Castle Liblice, Sept. 20-21, 1993.

J. Pollert, J. Zakin, J. Myska, P. Kratochvil, "Use of Friction Reducing Additives in District Heating System Field Test at Kladno – Krocehlavy, Czech Republic," International District Heat and Cooling Association Annual Meeting, Seatle, June 1994.

Z. Chara, J.L. Zakin, J. Myska, M. Severa, "The Effect of a Micellar Friction Reducing Additive on Turbulent Structure in a Pipe," Fifth European Turbulence Conference, Siena, Italy, July 1994.

P. Stern, J. Myska, B. Lu, B.C. Smith, L.C. Chou, J.L. Zakin, "Relationship of Flow Birefringence and Normal Stresses of Cationic Surfactant Systems to Their Turbulent Friction Reduction Characteristics," Fourth European Rheology Congress, Seville, Sept. 4-9, 1994.

J. Pollert, P. Komrzy and J. Pollert "Surfactant Drag Reducers for Reducing Energy Demands in District Heating Systems," New Requirements for Structures and Their Reliability, Prague, 1994.

J. Myska, J.L. Zakin and Z. Chara, "Viscoelasticity of a Surfactant and Its Drag Reducing Ability," Euromech Colloquium on Drag Reduction, Ravello, Italy, April 19-21, 1995.

J. Pollert, K. Svejkovsky, P. Komrzy and J.L. Zakin, "Flows of Surfactant Drag Reducers for Decreasing of Pumping Energy Demand in District Heating Systems," XXXIIth Congress of the International Association of Hydraulic Research, London, Sept. 11-15, 1995.

J. Myska, "SNIZENI T LAKOVYCH ZTRAT OTONPNYCH SOUSTAVACH, poster presented at Trade Fair in Brno, Sept. 11-17, 1995.

J. Myska,, Z. Chara, M. Severa, B. Lu, J.L. Zakin, "Turbulence Behavior of Highly Drag Reducing Surfactant Systems in Pipe Flow," Annual Meeting of American Institute of Chemical Engineers, Miami, Nov. 12-17, 1995.

Z. Chara, J.L. Zakin, J. Myska, M. Severa, "Turbulence Structures in a Pipe of Drag Reducing Cationic Surfactant Solutions," Eighth Beer-Sheva International Symposium on MHD Flows and Turbulence, Jerusalem, Israel, Feb. 25-29, 1996.

J. Myska, J.L. Zakin, "Comparison of Flow Behavior of Polymeric and Cationic Surfactant Drag Reducers," ASME Fluids Engineering Division Symosium on Turbulence Modification and Drag Reduction, San Diego, July 7-11, 1996.

J. Pollert, P. Komrzy, K. Svejkovsky, J. Pollert, Jr., B. Lu, J.L. Zakin, "Drag Reduction and Heat Transfer of Cationic Surfactant Solutions," ASME Fluids Engineering Division Sysmposium on Turbulence Modification and Drag Reduction, San Diego, July 7-11, 1996.

J.L. Zakin, J. Myska, Z. Lin, "Similarities and Differences in Drag Reduction Behavior of High Polymer and Surfactant Solutions," International Symposium on Seawater Drag Reduction, Newport, RI, July 22-24, 1998.

J.L. Zakin, J. Myska, Z. Lin, "Some Surprising Behaviors of Surfactant Drag Reducing Systmes," 11th European Drag Reduction Working Meeting, Sept. 15-17, 1999, Prague — Plenary Lecture.

Y. Qi, Y. Kawaguchi, Z. Lin, M. Ewing, R.N. Christensen, J.L. Zakin, "Enhancing Heat Transfer in Drag Reducing Surfactant Solutions," 11th European Drag Reduction Working Party Meeting, Sept. 15-17, 1999, Prague.

J. Myska, Z. Chara, "A Puzzle in the Investigation of Drag Reduction Caused by Construction of the Experimental Stand," 11th European Drag Reduction Working Party Meeting, Sept. 15-17, 1999, Prague.

J. Myska, Z. Chara, "New View of Surfactant Drag Reduction Ability," 11th European Drag Reduction Working Party Meeting, Sept. 15-17, 1999, Prague.

*Note*: Underlined author presented the paper.

## Publications

Z. Chara, J.L. Zakin, M. Severa, J. Myska, "Turbulence Measurements of Drag Reducing Surfactant Systems," Experiments in Fluids, 16, 36-41 (1993).

J.L. Zakin, L.C. Chou, B.C. Smith, B. Lu, "Molecular Variables Affecting Drag Reduction by Cationic Surfactants," Proc. Int. Conf. On Fluid Mechanics and Hydrodynamical Aspects of Biosphere, Academy of Sciences of Czech Republic, Liblice, pp 20-25, September 1993.

J. Pollert, P. Komzry, A. Vozenilek, J.L. Zakin, "Influence of Pipe Diameter and Temperature on Efficiency of Drag Reducing Surfactants," Proc Proc. Int. Conf. On Fluid Mechanics and Hydrodynamical Aspects of Biosphere, Academy of Sciences of Czech Republic, Liblice, pp 61-67, September 1993.

J. Myska, M. Severa, J.L. Zakin, "LDA Investigation of Turbulent Characteristics of Drag Reducing Surfactant Systems," Proc. Int. Conf. On Fluid Mechanics and Hydrodynamical Aspects of Biosphere, Academy of Sciences of Czech Republic, Liblice, pp 73-79, September 1993.

Z. Chara, J. Myska, M. Severa, J.L. Zakin, ""LDA Investigation of Turbulent Characteristics of Drag Reducing Surfactant Systems," J. Hydrol. Hydromech., 42, 141-150, 1994.

J. Pollert, J.L. Zakin, J. Myska, P. Kratochvil, "Use of Friction Reducing Additives in District Heating System Field Test at Kladno – Krocehlavy, Czech Republic," Proc. International District Heat and Cooling Association Annual Meeting, Seatle, Vol 85, 141-156, 1994.

J. Pollert, P. Komrzy, J. Pollert Jr., " Surfactant Drag Reducers for Reducing Energy Demands in District Heating Systems," Proc. New Requirements for Structures and Their Reliability, pp. 171-176, Prague, 1994.

P. Stern, J. Myska, B. Lu, B.C. Smith, L.C. Chou, J.L. Zakin, "Relationship of Flow Birefringence and Normal Stresses of Cationic Surfactant Systems to Their Turbulent Friction Reduction Characteristics," Proc. Fourth European Rheology Congress, pp. 605-608, Seville, Sept. 4-9, 1994.

J. Myska and P. Stern, "Properties of a Drag Reducing Micelle System," Colloid and Polymer Science, 272, 642-647, 1994.

J. Pollert, K. Svejkovsky, P. Komrzy and J.L. Zakin, "Flows of Surfactant Drag Reducers for Decreasing of Pumping Energy Demand in District Heating Systems," Proc. XXXIIth Congress of the International Association of Hydraulic Research, London, Sept. 11-15, 1995 in Part 2: Industrial Hydraulics and Multi-Phase Flows, pp. 1152-1158.

J. Myska, Z. Chara, J.L. Zakin, "Viscoelasticity of a Surfactant and its Drag Reducing Ability," Appl. Sci Research, 55, 297-310, 1996.

J. Pollert, P. Komrzy, K. Svejkovsky, J. Pollert, Jr., B. Lu, J.L. Zakin, "Drag Reduction and Heat Transfer of Cationic Surfactant Solutions," Proc. ASME Fluids Engineering Division Symposium on Turbulence Modification and Drag Reduction, ASME pub FED-vol 237, pp. 31-36, 1996.

J. Myska, J.L. Zakin, "Comparison of Flow Behavior of Polymeric and Cationic Surfactant Drag Reducers," Proc. Proc. ASME Fluids Engineering Division Symposium on Turbulence Modification and Drag Reduction, ASME pub FED-vol 237, pp. 165-168, 1996.

J.L. Zakin, J. Myska, Z. Chara, "New Limiting Drag Reduction and Velocity Profile Asymtotes for Polymeric Additive Systems," AIChE J., 42, 3544-3546, 1996.

J. Myska, P. Stepanek, J.L. Zakin, "Micellar Size of Drag Reducing Cationic Surfactants," Colloid & Polymer Science, 275(3), 254-262, 1997.

J. Myska, J.L. Zakin, "Difference in the Flow Behaviors of Polymeric and Cationic Surfactant Drag Reducing Additives," Ind. Eng. Chem. Research, 36, 5483-5487, 1997.

J. Myska, Z. Chara, J.L. Zakin, " Viscoelasticity of an Additive Used as a Drag Reducer in a District Heating System," J. Hydrol. Hydromech, 45, 151-172, 1997.

J.L. Zakin, J. Myska and Z. Lin, "Similarities and Differences in Drag Reduction Behavior of High Polymer and Surfactant Solutions," Proc. Int. Symposium on Seawater Drag Reduction, pp. 277-280, Newport, RI, July 1998.

J.L. Zakin, J. Myska, Z. Lin, "Some Surprising Behaviors of Surfactant Drag Reduction Systmes," Proc. 11th European Drag Reduction Working Party Meeting, pp. 5, Prague, Czech Republic, September 15-17, 1999.

Y. Qi, Y. Kawaguchi, Z. Lin, M. Ewing, R.N. Christensen, J.L. Zakin, "Enhanced Heat Transfer of Drag Reducing Surfactant Solutions with Fluted Tube-in-Tube Heat Exchanger," Proc. 11th European Drag Reduction Working Party Meeting, pp. 42, Prague, Czech Republic, September 15-17, 1999.

J. Myska and Z. Chara, "New View of Surfactant Drag Reduction Ability," 11th European Drag Reduction Working Meeting, pp. 32-33, Prague, Czech Republic, September 15-17, 1999.

J. Myska, Z. Chara, "A Puzzle in the Investigation of Drag Reduction Caused by Construction of the Experimental Staud," 11th European Drag Reduction Working Meeting, pp. 34, Prague, Czech Republic, September 15-17, 1999.

Y. Qi, Y. Kawaguchi, Z. Lin, M. Ewing, R.N. Christensen, J.L. Zakin, "Enhancing Heat Transfer in Drag Reducing Surfactant Solutions," Int. J. Heat and Mass Transfer, in press.

## VI. Project Productivity

The Project's research, personal and infrastructure objectives are described in II. All of the objectives were met.

## VII. Future Work

Dr. Myska, Chara and I have submitted a Collaborative Linkage Grant proposal to NATO to continue collaborative research on improved drag reducing additives and their rheological behavior. The Institute of Hydrodynamics is funding another district heating field test in one of

their research buildings. The new Akzo Nobel zwitterionic/anionic drag reducing additive will be used in the test to be conducted in winter 2000-2001.

## VIII. Literature Cited

The publications resulting from this project are listed in Section V—Publications. They contain a large number of cited papers. A specific paper cited in this report is: (1) P. Virk, "Drag Reduction Fundamentals," AIChE J, 21, 625-656 (1975).

Experiments in Fluids 16, 36–41 (1993)

Experiments in Fluids

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# Turbulence measurements of drag reducing surfactant systems

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**Abstract.** LDA measurements were made of mean velocity and of turbulence intensity in a 39.4 mm diameter tube, the first measurements in three directions on drag reducing surfactant solutions (0.05% and 0.1% Habon G). Drag reduction exceeded the predictions of the Virk maximum drag reduction asymptote and elastic sublayer mean velocity profiles are steeper than the profile proposed by Virk for maximum drag reducing asymptote solutions. Axial turbulence intensities for Habon G solutions are higher than those for water near the wall, lower in most of the outer region and about the same at the center. Tangential and radial turbulence intensities are lower than those for water.

**List of symbols**

| | |
|---|---|
| $D$ | tube diameter |
| $f$ | friction factor |
| $\frac{\Delta P}{l}$ | pressure gradient |
| $Re$ | Reynolds number |
| $r$ | distance from tube center |
| $R$ | tube radius |
| $u'_z$, $u'_r$ and $u'_\varphi$ | fluctuating velocity in the axial, radial and tangential directions |
| $u_z$ | mean local axial velocity |
| $u_{max}$ | mean velocity at tube center |
| $\bar{u}$ | bulk mean velocity |
| $u^+$ | dimensionless velocity |
| $u^*$ | friction velocity |
| $x$ | distance from the wall |
| $y^+$ | dimensionless distance from the wall |
| $\rho$ | density |
| $\delta$ | thickness of wall layer |
| $\eta$ | viscosity |
| $\tau_w$ | wall shear stress |
| subscript $s$ | refers to surfactant solution flow |
| subscript $w$ | refers to water flow |

A version of this paper was presented at the 13th Symposium on Turbulence, University of Missouri-Rolla, Sept. 21–23, 1992

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## 1 Introduction

Considerable research has been carried out on the influence of high polymers on drag reduction in turbulent flow in pipes. A major restriction on the use of polymer drag reducers is their susceptibility to mechanical degradation which limits their usefulness in circulation systems. Friction losses can also be significantly reduced by the use of certain cationic, anionic and non-ionic surfactants. Sufactant drag reducers are also degraded under high shear but quickly regain their drag reduction effectiveness when flowing in regions of low shear. Thus, they can be used in recirculation flows such as in district heating or district cooling systems. The great disadvantage of surfactants is their disposition to create rich foam, however when free surfaces are exposed.

The surfactant additive which is being widely considered for use in district heating systems is Habon G, a product of the Hoechst company, which is designed for use at temperatures of 30 to 100°C. Accordingly a study of the turbulent flow characteristics of this surfactant has been undertaken. In this paper, LDA measurements of mean velocity profiles and of turbulence intensities in all three directions in pipe flow are reported for highly drag reducing solutions of this additive, the first turbulence measurements in all three directions on a drag reducing surfactant system.

## 2 Experimental

### 2.1 Hydraulic measurement stand

The facility for LDA measurements of turbulent flow is shown in Fig. 1. Water is kept at a constant level in an overflow tank (*3*) by an over flow tube (*4*) which can be vertically shifted. The glass tube (*8*) of inner diameter 39.4 mm is 5.80 m long. The test section is approximately

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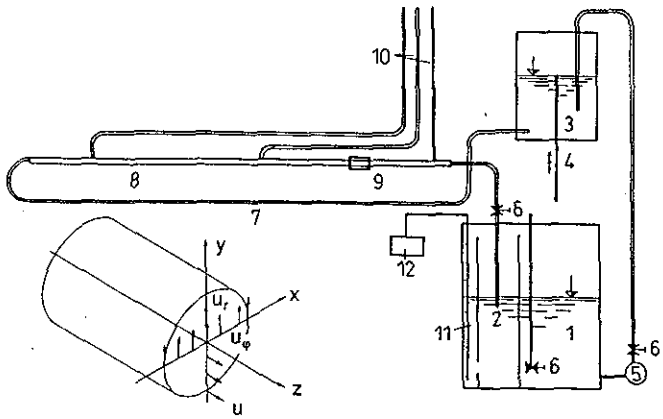



**Fig. 1.** Hydraulic stand for LDA measurements with glass tube of 39.4 mm inner diameter and schematic diagram of velocity components

2 m long and it is provided with open manometers (*10*) for measuring pressure drops. A rectangular box (*9*) made of thin glass envelope the tube in the middle of the test section. The box is filled with water in order to suppress undesired reflections of the laser beams. Level gauge (*11*) connected with electric timer (*12*) is installed in a separate calming chamber of the collecting tank (*1*) in order to measure the rate of flow. Velocity fluctuations, $u'_z$, $u'_r$ and $u'_\varphi$, and mean local velocity, $u_z$, could be measured. LDA measurements were made at a location 4.2 m downstream from the entrance of the tube section.

### 2.2 *LDA system*

The LDA optics are mounted together with the laser on a remote controlled *xyz* support. The Carl Zeiss Jena (FRG) ILA 120-1 Argon ion laser operates with a single wave length ($\lambda = 488$ nm) at the fundamental mode $TEM_\infty$. The laser beam enters the DISA (Denmark) 55 L 83 beam splitter and Bragg cell section and is split into two parallel beams, one of which is frequency shifted by 40 $MH_z$. The laser beams are then directed by a mirror into the lens. The lens focuses the laser beams into a mutual intersection, which is the optical probe of the LDA. The actual length of the optical probe is 0.8 mm and the diameter is 0.3 mm.

The differential Doppler signal is detected by the Dantec (Denmark) photomultiplier section 55 × 08 with PM optics 55 × 34 and transformed into an electrical signal for further electronic processing. The evaluation of the Doppler signal was performed by a Dantec Particle Dynamics Analyzer, which consists of the PDA signal processor and computer COMPAQ DESKPRO 386s.

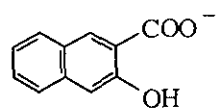
On this processor only six different bandwidths can be selected (0.12; 0.4; 1.2; 4; 12 and 36 MHz). For the longitudinal velocities (both in water and in Habon G solution) the effective range of −1 to +3 MHz was chosen and for cross fluctuation measurements the effective range from −0.3 to +0.9 MHz was used.

### 2.3 *Surfactant system*

The surfactant used was Habon G, a product of the Hoechst Company, FRG. The content of delivered material is 53.5% active surfactant, 10.2% isopropanol and 36.3% water. Concentrations reported are the concentrations of active surfactant. The cation of the surfactant is hexadecyldimethyl hydroxyethyl ammonium

$$[C_{16}H_{33}N(CH_3)_2C_2H_4OH]^+$$

and the counter-ion is 3-hydroxy-2-naphthoate




The surfactant molecules form large rod-shaped micelles which are effective in causing drag reduction (Bewersdorff and Ohlendorf 1988).

The surfactant material is a dark brown liquid of high viscosity which is easily diluted and becomes an opalescent whitish liquid when dispersed in water. At concentrations above 0.1 − 0.2% the solution loses its transparency when flowing in a 40 mm glass tube and so LDA measurements at concentrations above 0.1% are not possible. The foam formed on the liquid surface is white, rich and dense.

## 3 Results and discussion

Habon G is a leading candidate for use as a drag reducer in district heating systems and was reported to be effective at temperatures of 30–100°C by Fankhänel (1991) and Althaus (1991). The data in Fig. 2 show that Habon G is also highly effective as a drag reducer at room temperatures, i.e. 18–26°C. There is no difference in efficiency at concentrations of 0.107% and 0.053%. Drag reduction for the 0.053% and 0.107% solutions was greater than that predicted by the Virk (1975) maximum drag reduction asymptote or D.R.A.:

$$\frac{1}{\sqrt{f}} = 19\log_{10} \; Re\sqrt{f} - 32.4 \qquad (2)$$

where $f = \dfrac{\Delta PD}{2l\rho\bar{u}^2}$ and $Re = \dfrac{D\bar{u}\rho}{\eta}$

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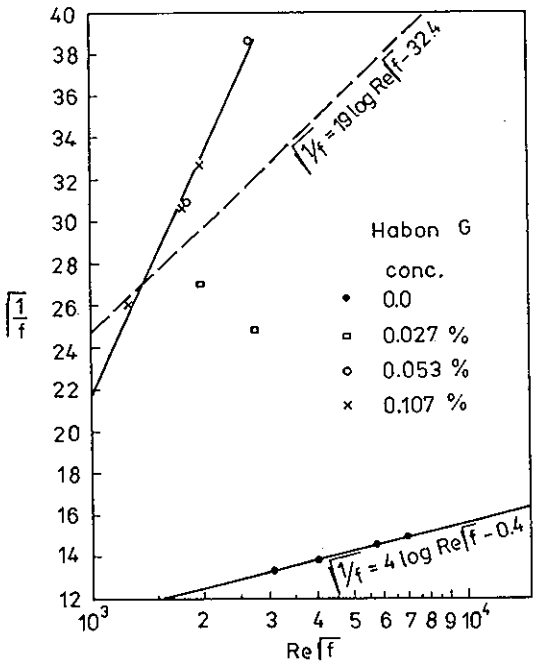


Fig. 2. Drag reducing efficiency of Habon G solutions in Prandtl-Karman coordinates at room temperature

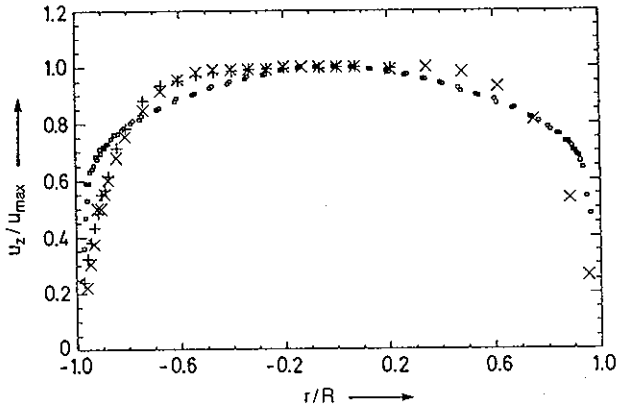


Fig. 3. Profile of mean local axial, velocity, $u_z$ divided by maximum velocity, $u_{max}$. □ water, $Re=104{,}600$ and $Re=44{,}500$; + Habon G solution, conc. 0.053%, $Re=113{,}880$; $X$ Habon G solution, conc. 0.107%, $Re=39{,}280$

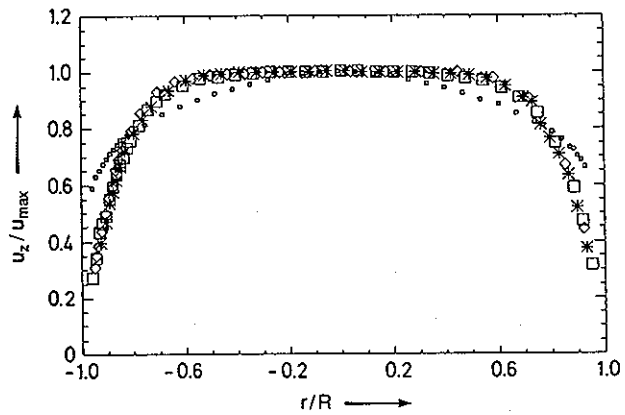


Fig. 4. Profile of mean local axial velocity, $u_z$, divided by maximum velocity, $u_{max}$. □ water, $Re=104{,}600$, □ Habon G solution, conc. 0.107%, $Re=78{,}560$; * Habon G solution, conc. 0.107%, $Re=57{,}740$; ◇ Habon G solution, conc. 0.053%, $Re=57{,}860$

Viscosities of the surfactant solutions after degradation by the centrifugal pump differ from water viscosity by less than 5%.

At a concentration of 0.027%, however, the efficiency is greatly reduced at these low temperatures. In the turbulence measurements for the 0.053% and 0.107% solutions, drag reduction was 73% at the smallest Reynolds number and 84% at the greatest Reynolds number.

Figures 3 and 4 show the velocity profiles obtained with water solutions of Habon G at concentrations of 0.053% and 0.107% in comparison with velocity profiles of water. The plots show distribution of the mean local axial velocity, $u_z$, normalized by the maximum velocity at the tube axis, $u_{max}$, vs dimensionless tube radius, $r/R$. The Reynolds numbers were 39,280, 57,860, 78,480 and 113,880. Velocity profiles for the two Habon G concentrations are similar indicating no significant effect of the surfactant concentration on the velocity profile in this concentration range just as there was no effect on their drag reduction behavior (Fig. 2). The values of $u_z/u_{max}$ of Habon G solutions are quite flat in the central region. They are higher than those of water in most of the core region and they are smaller in the wall region. They would be expected to become more convex and approach the water velocity profiles at much higher wall shear stresses as the micelle network breaks up and drag reduction is lost. The thickness of the wall layer for the water experiments can be estimated from the semi-empirical Karman theory, $\delta=32.5\ D/Re\sqrt{4f}$ (see Smetana 1957). For $Re=44{,}500$, $\delta=0.19$ mm and for $Re=104{,}600$, $\delta=0.09$ mm, with about twice these values for Habon G solutions at similar Reynolds numbers. It was not possible to measure this close to the wall; measurements could be made only as close as about 0.5 mm to the wall. The velocity profiles for water superimpose as do those for the Habon solutions. The data also nearly superimpose when plotted as $u^+$ vs log $y^+$ as can be seen in Fig. 5.

$$u^+ = \frac{u_z}{u_*} \quad \text{and} \quad y^+ = \frac{xu^*\rho}{\eta} \tag{3}$$

where $u^* =$ friction velocity $= \sqrt{\tau_w/\rho}$

$\tau_w =$ wall shear stress

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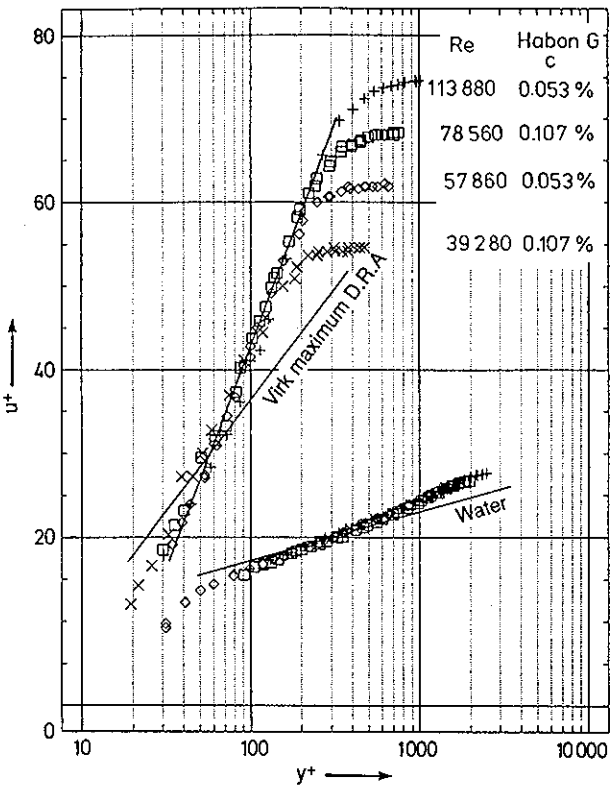


Fig. 5. Dimensionless velocity profiles, $u^+ - \log_{10} y^+$ in water and in Habon G solutions

For all Reynolds numbers measured, the $u^+$ vs $y^+$ data for water lie in a narrow band, see Fig. 5. The slope is a little steeper than the universal equation based on Nikuradse's measurements as reported by Schlichting (1960).

$$u^+ = 5.75 \log_{10} y^+ + 5.5 \tag{4}$$

Generally, our measured point velocities, $u_z$, are slightly greater than predictions from Nikuradse's equation in the center of the tube and lower nearer the wall. We have not yet been able to reconcile this discrepancy but we note that similar deviations from this equation were observed by several authors as reported by Monin and Iaglom (1971).

The equation fitting the data in the elastic sublayer region is:

$$u^+ = 53.9 \log_{10} y^+ - 65 \tag{4}$$

At a certain value of $y^+$, the function $u^+ = f(y^+)$ deviates from this equation and the function becomes nearly horizontal in the flat profile center region. This value of $y^+$ increases with *Re*. The slope of the common profile line is greater than that proposed by Virk (1975) for the elastic region:

$$u^+ = 26.9 \log_{10} y^+ - 17 \tag{5}$$

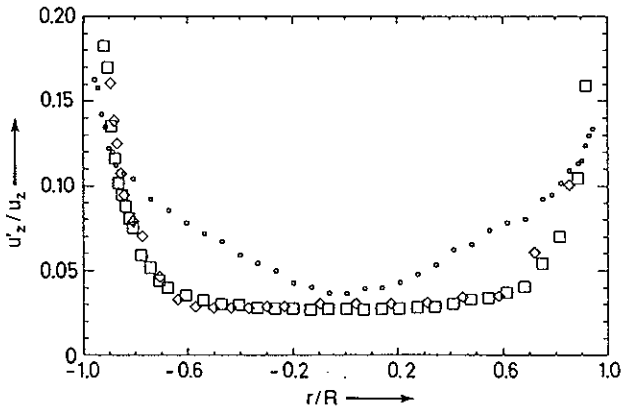


Fig. 6. Profiles of axial turbulence intensities normalized by mean local axial velocity. □ water, $Re = 104{,}600$; □ Habon G solution, conc. 0.107%, $Re = 78{,}560$; ◇ Habon G solution, conc. 0.053%, $Re = 57{,}860$

It is closer to the value of 44.9 reported by Bewersdorff and Thiel (1993).

The S-shape profiles with slopes steeper than Eq. 5 shown in Fig. 5 resemble other velocity profiles in the literature for systems showing large drag reduction. For example, data of Bewersdorff and Ohlendorf (1988) for tetradecyl trimethyl ammonium salicylate (TTAS) have this shape with sections steeper than Eq. 5. Those authors also cite several Russian papers by Povkh and coworkers which showed similar profile shapes for surfactant solutions (see for example Povkh et al. 1988). A polymer solution example is data of Schümmer and Thielen (1980) who reported data for high molecular weight polyacrylamide solutions showing large drag reduction and whose velocity profiles are also S-shaped and steeper than the prediction of Eq. 5.

Profiles of velocity fluctuations in the *z* direction were also measured for water and for surfactant solutions (Fig. 6). The water results are similar to those of Patterson (1966) for benzene at *Re* of 82,615 in a 50.8 mm tube. The ratio of root mean square longitudinal fluctuations to the mean local velocity, $u'_z/u_z$, is generally smaller for the micelle systems in most of the cross section but the values are almost the same at the tube center. They exceed the values for water in the wall region where the mean velocities of the micelle system are lower than those for water. When plotted as $u'_z/u^*$, they are much higher than water intensities because $u^*$ is relatively small for these highly drag reducing systems (Fig. 7). Intensities above eight were observed. Peak values near eight were also observed by Bewersdorff and Ohlendorf (1988) for a TTAS system with high drag reduction.

Micelles greatly reduce the cross fluctuations. The root mean square tangential velocity fluctuations, $u'_\varphi$, are normalized by the average cross-section velocity, $\bar{u}$, both in the surfactant system, $u'_\phi/\bar{u}_s$, and in water, $u'_{\varphi w}/\bar{u}_w$, and

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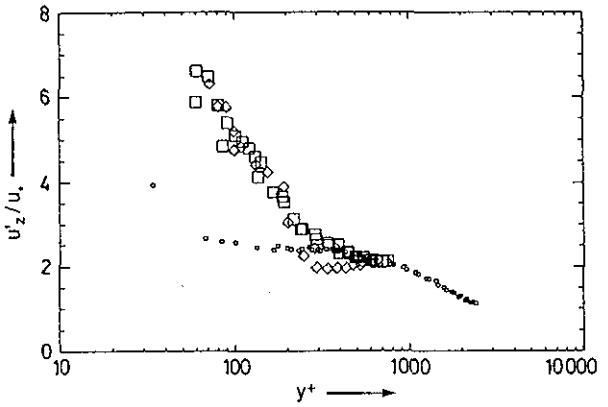


Fig. 7. Profiles of axial turbulence intensities normalized by friction velocity. □ water, $Re = 104{,}600$; □ Habon G solution, conc. 0.107%, $Re = 78{,}560$; ◇ Habon G solution, conc. 0.053%, $Re = 57{,}860$

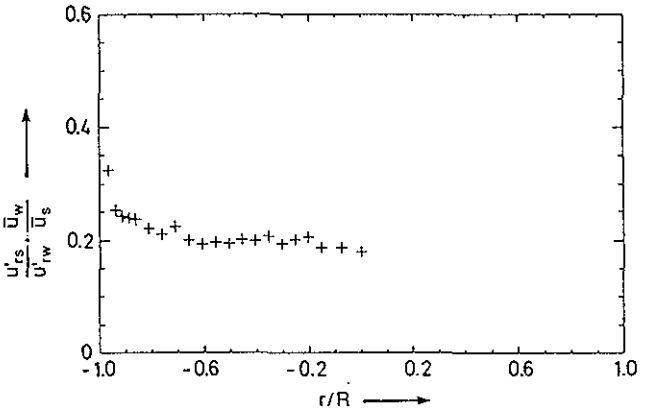


Fig. 9. Profile of ratio of tangential velocity fluctuations in surfactant to that in water normalized by the ratio of bulk mean velocity of surfactant and of water. + Habon G solution, conc. 0.053%, $Re = 57{,}860$

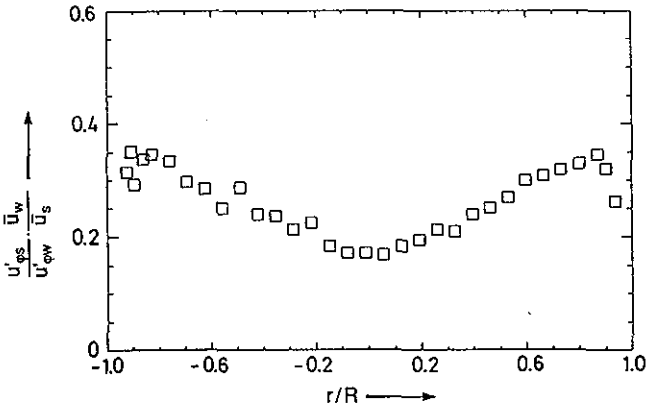


Fig. 8. Profile of the ratio of tangential velocity fluctuations in surfactant to that in water normalized by the ratio of bulk mean velocity of surfactant and of water. □ Habon G solution, conc. 0.053%, $Re = 57{,}860$

a profile of

$$\frac{u'_{\varphi s}}{\bar{u}_s} \Big/ \frac{u'_{\varphi w}}{\bar{u}_w} = \frac{u'_{\varphi s}}{u'_{\varphi w}} \cdot \frac{\bar{u}_w}{\bar{u}_s} \tag{6}$$

is plotted in Fig. 8. Nearly equal Reynolds numbers with nearly equal average velocities were chosen for evaluation. The micelle secondary network in the stream reduces the tangential fluctuations to 20% (center) and 35% (closer to the wall) of those in water. At $r/R$ values greater than 0.85 the ratio begins to decrease rapidly.

Even greater damping of cross fluctuations can be found with root mean square radial fluctuations, $u'_r$. Normalization is done in the same way as above,

$$\frac{u'_{rs}}{\bar{u}_s} \Big/ \frac{u'_{rw}}{\bar{u}_w} = \frac{u'_{rs}}{u'_{rw}} \cdot \frac{\bar{u}_w}{\bar{u}_s} \tag{7}$$

with the same average velocities. The data are shown in Fig. 9. The whole profile shows 80% damping of radial fluctuations, $u'_{rs}$, in the micelle system compared with $u'_{rw}$. Bewersdorff (1990) reports ratios of $u'_{rs}$ to $u'_{zs}$ of 0.39 to 0.47 for a TTAS drag reducing solution which are smaller than our values of $\approx 0.6$. It is interesting that the ratio, $u'_{rs}/u'_{rw}$ increases in the wall region while the ratio $u'_{\varphi s}/u'_{\varphi w}$ appears to decrease in this region. Measurements closer to the wall are needed to clearly define these trends.

It should be stressed in conclusion that for highly drag reducing surfactant systems fluctuations of the radial and tangential velocity components are damped significantly compared to water flows while fluctuations of the longitudinal velocity component in the wall region are enhanced.

## 4 Conclusions

1. Cationic surfactant solutions in water can reduce turbulent friction losses more than predicted by the Virk maximum drag reduction asymptote.
2. An elastic sublayer mean velocity profile in highly drag reducing surfactant solutions is steeper than take profile proposed by Virk as the maximum drag reducing asymptote for polymer solutions.
3. The mean velocity profiles observed for drag reducing surfactant systems are flatter than most of those reported for polymer solutions in the core region.
4. Absolute values of axial turbulence intensities for drag reducing surfactant systems near the wall are higher than those for water, lower than water in most of the outer region and about the same at the center of the tube when compared at similar *Re*. They are much higher than water, except at the center, when reduced by the friction velocity, $u'_z/u^*$.

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5. Tangential turbulence intensities for drag reducing surfactant systems are reduced to 20 to 35% of those for water at all locations in the tube.
6. Radial turbulence intensities for drag reducing surfactant systems are reduced to about 20% of those for water in most locations in the tube but rise somewhat close to the wall.

## Acknowledgements

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J. Hydrol. Hydromech., *42*, 1994, 2–3, 141—150

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# LDA INVESTIGATION OF TURBULENT CHARACTERISTICS OF DRAG REDUCING SURFACTANT SYSTEMS

ZDENĚK CHÁRA, JIŘÍ MYŠKA, MIROSLAV SEVERA, JACQUES L. ZAKIN

The paper describes the results of an experimental investigation of turbulent structure in two drag reducing surfactant systems – HABON G and ETHOQUAD T/13. The profiles of the local velocities as well as the intensities of turbulence were measured in a horizontal glass pipe with an inner diameter of 39.4 mm.

KEY WORDS: Drag-Reduction, Surfactants, Turbulence.

Zdeněk Chára, Jiří Myška, Miroslav Severa, Jacques L. Zakin: Měření turbulentních charakteristik micelárních látek snižujících tření metodou LDA. Vodohosp. Čas., *42*, 1994, 2–3; 3 lit., 5 obr.

Příspěvek se zabývá měřením turbulentních charakteristik dvou povrchově aktivních aditiv: HABON G A ETHOQUAD T/13. Profily bodových rychlostí a intenzit turbulence byly měřeny na skleněném horizontálním kruhovém potrubí s vnitřním průměrem 39,4 mm.

KLÍČOVÁ SLOVA: snižování tření, aditiva, turbulence.

Зденек Хара, Иржи Мышка, Мирослав Севера, Джек Л. Закин: Измерение турбулентных характеристик мицеларных веществ, понижающих гидродинамическое сопротивление с помощью ЛДА. Водохозяй. Ж., *42*, 1994, 2–3; 3 лит., 5 рис.

Описывается измерение турбулентных характеристик водных растворов двух поверхно активных веществ: HABON G и ETHOQUAD T/13. Были измерены распределения точечных скоростей и интенсивностей турбуленции тока раствора в стекляной горизонтальной трубе внутренного диаметра 39.4 мм.

КЛЮЧЕВЫЕ СЛОВА: понижение сопротивления, поверхно активные вещества, турбуленция.

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## 1. Introduction

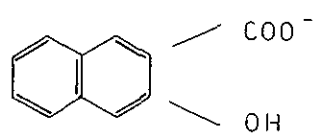
The use of drag reducing additives in district heating system has stimulated interest in surfactant friction reducers over the past few years. While the applicability of high polymer additives in circulation systems is limited by their susceptibility to mechanical degradation and loss of effectiveness, surfactant drag-reducers appear to be more suitable. Although they may also suffer mechanical degradation in regions of high shear, they regain their drag reducing effectiveness when the level of the shear stress decreases.

## 2. Experimental part

For detailed investigation of turbulent structure in surfactant systems, we have chosen two commercial surfactants HABON G and ETHOQUAD T13/50. because of their high drag reducing efficiency at room temperatures. Habon G is the product of HOECHST Comp., FRG. The content of delivered material is 53.5% active surfactant, 10.2% isopropanol and 36.3% water. The cation of the surfactant is hexadecyl-trimethyl-ammonium

$$[C_{16}H_{33}N(CH_3)_2C_2H_4OH]^+$$

and the counter-ion is 3-hydroxy-2-naphthoate




The surfactant material is a dark brown liquid with high viscosity which is easily diluted in water.

Ethoquad T13/50 (ET) is a product of the AKZO CHEMIE AMERICA, USA. The content of the delivered material is 50% active surfactant, 36% isopropanol and 14% water. The surfactant is tallow tris hydroxyethyl ammonium acetate

$$\text{Tallow} - N\,(C_2H_4OH)_3\ Ac-$$

with molecular weight $M_w = 454$. This surfactant must be mixed with natrium salicylate (NA) in order to exhibit drag-reducing properties.

High-temperature pressure-loss experiments were carried out in a small recirculation pumping system containing a smooth stainless steel pipe with inner diameter of 6.17 mm. Pressure drops were measured by a Validyne differential pressure transducer and flow rates were measured with a Rosemount Series 8700



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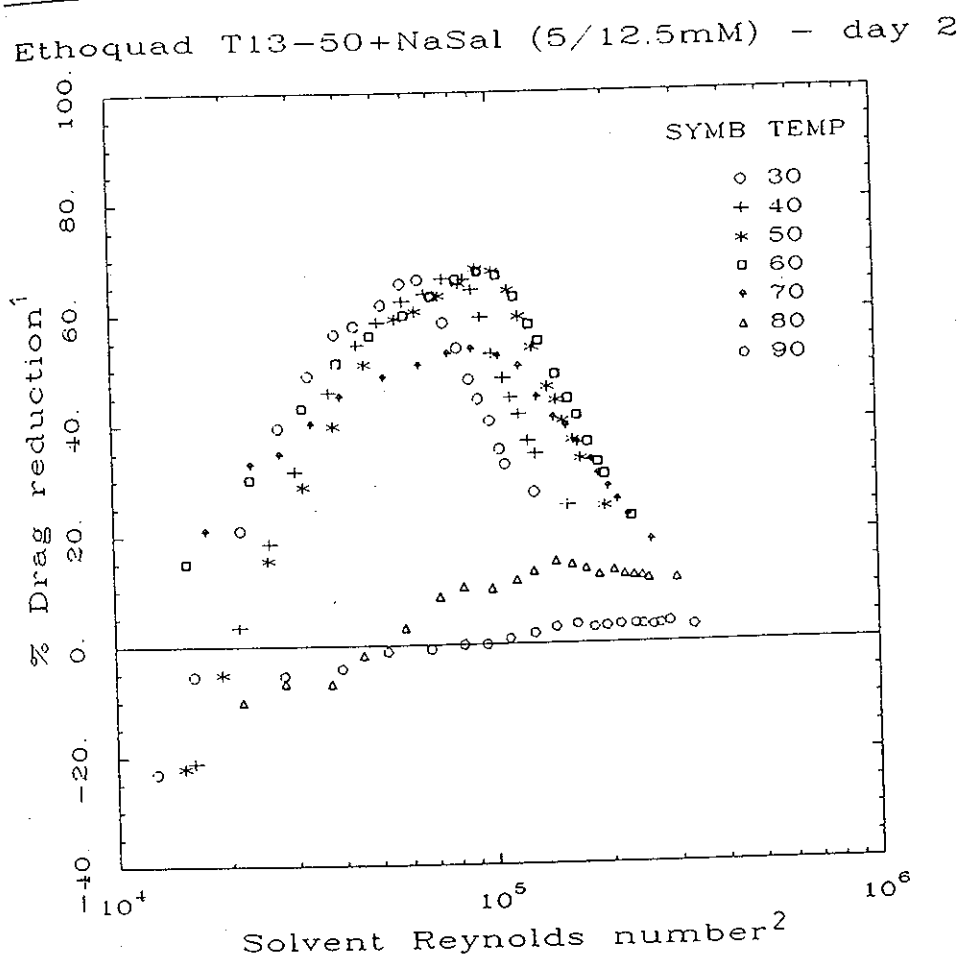


Fig. 1. Drag reducing efficiency of ET/NA solution for the temperature range between 30° and 90°C (results from a 6.17 mm stainless steel pipe).
Obr. 1. Účinnost snižování tření pro roztok ET/NA a pro rozsah teplot od 30° do 90°C (měření v 6.17 mm nerez potrubí); 1 – % snižování tření, 2 – Reynoldsovo číslo.

magnetic flow meter. The pump was an Oberdorfe bronze rotary gear pump driven by a variable speed drive. High temperature runs (up to 130°C) were carried out using two 2000 watt heaters along with a 750 watt trimmer heater which was connected to a regulator which could control the temperature to ±1°C. The results for Ethoquad T/13 with a concentration of 0.654% of the total weight (ET 5 mMol/l active surfactant and NA 12 mMol/l) are plotted in Fig. 1, where good drag efficiency can be seen for the temperature range between 30°C and 70°C. LDA measurements of mean velocities and turbulence intensities were carried out using



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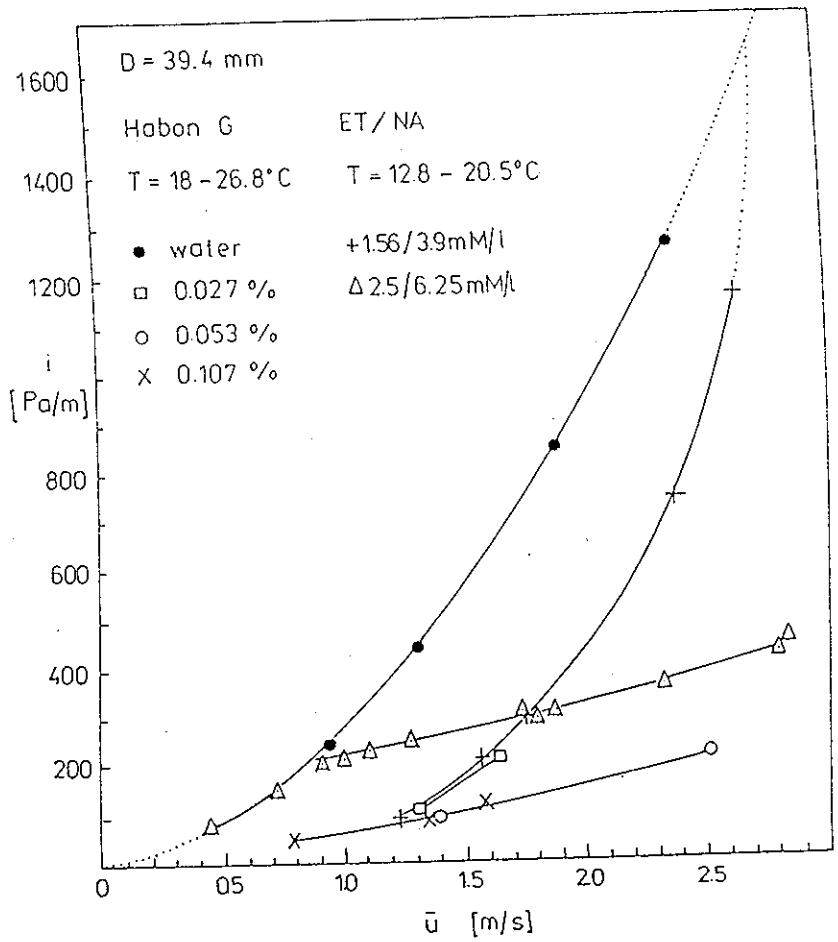


Fig. 2. The efficiency of HABON G and ET/NA solutions in drag reduction in a glas tube with diameter of 39.4 mm (total surfactant weight concentration 0.204% is ET/NA =1.56/3.9 mMol/l and 0.327% is ET/NA=2.50/6.25 mMol/l of active surfactant, as fo Habon G weight concentration 0.053% equals 1.06 mMol/l of active surfactant).
Obr. 2. Účinnost snižování tření pro roztoky HABON G a ET/NA ve skleněném potru o průměru 39.4 mm (váhová koncentrace 0,204 % je ET/NA=1,56/3,9 mMol/l a 0,327 je ET/NA 2,50/6,25 mMol/l aktivního surfactantu a pro Habon G je váhová koncentra 0,053 % rovna 1,06 mMol/l aktivního surfactantu).

a facility with a closed recirculating loop where a glass tube with inner diameter 39.4 mm was installed [2]. The LDA optics were mounted together with the laser a remote-controlled xyz support. The Carl Zeiss Jena (FRG) ILA 120-1 Argon i laser which operates with a single wave length ($\lambda = 488$ nm) was used for Habon



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measurements and the Coherent INNOVA-70 Argon ion laser ($\lambda = 514.5$ nm) was used for ET/NA measurements. The differential Doppler signals were detected by the Dantec photomultiplier $55 \times 08$ in the forward scatter direction. The evaluation of the Doppler signals was performed using the Dantec PDA systems and a Dantec Counter processor.

## 3. Results

The data in Fig. 2 show the efficiency of the surfactants as drag reducers at room temperatures, i.e. 18–26°C for Habon-G and 12.8–20.5°C for ET/NA for various weight concentrations of active matter. There is no difference in the efficiency at weight concentrations of 0.107% and 0.053% of active matter of Habon-G. At a concentration of 0.027%, the efficiency is greatly reduced at these low temperatures. The solutions of ET/NA exhibit interesting behaviour: there is a certain region (below $\overline{u} = 1.7$ m/s) where the less concentrated 0.204% solution is more efficient than the more concentrated 0.327% solution. The picture also shows that, for comparable effect, a much greater amount of ET/NA is needed. The efficiency of both surfactants was measured at the beginning of the experiments and it was again checked after the experiments were finished. No change was observed.

The dimensionless velocity profiles ($u/u_{\max}$) in water and in both surfactants are plotted in Fig. 3. The velocity profiles of ET/NA (weight concentration 0.204%) are very similar to the profile in water (Fig. 3a), but for a concentration of ET/NA of 0.327% and for Habon G (concentration 0.053 % and 0.107%) the velocity profiles differ from the water profile (Fig. 3b), where flatter velocity profiles can be observed in the central region.

This fact is more obvious in Fig. 4, where the $u^+$ vs $y^+$ data are depicted. The equation fitting the data for the Habon G surfactant in the elastic sublayer region is

$$u^+ = 53.9 \log y^+ - 65 \tag{1}$$

and the equation for ET/NA (concentration 0.327%) is

$$u^+ = 53.9 \log y^+ - 84. \tag{2}$$

At a certain value of $y^+$, the function $u^+ = f(y^+)$ deviates from these equations and the function becomes nearly horizontal in the flat velocity profile region. The slope of the common profile line is greater than that proposed by Virk [3] for the elastic region

$$u^+ = 26.9 \log y^+ - 17. \tag{3}$$

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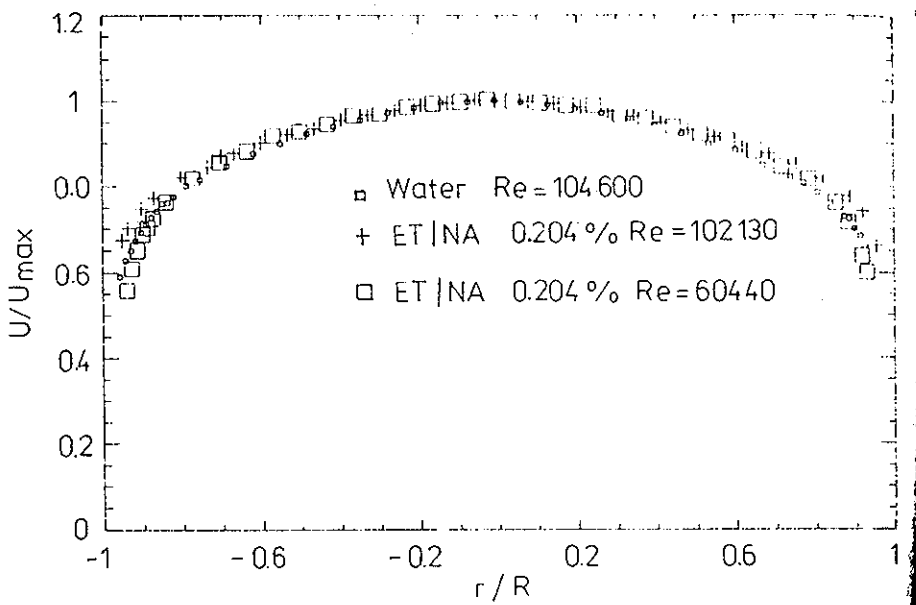


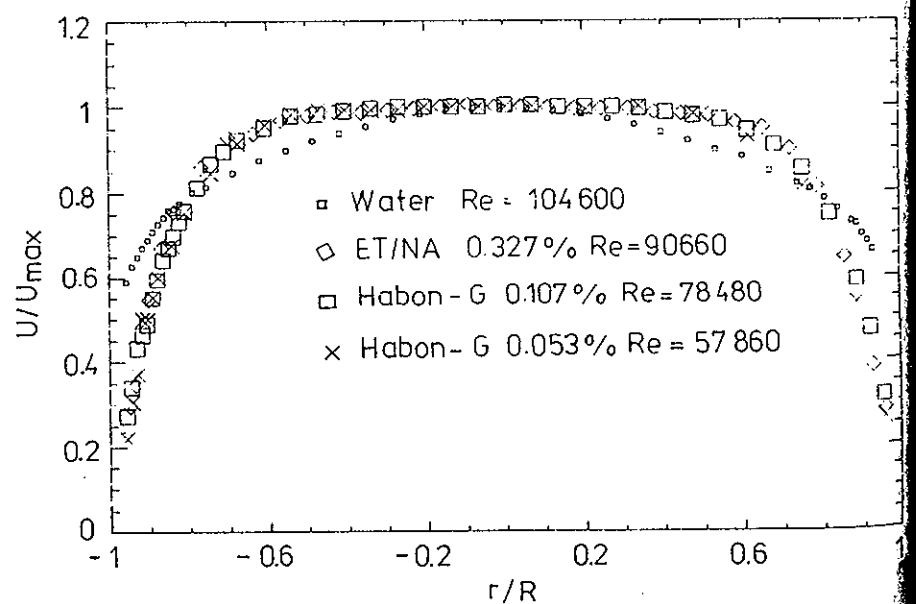


Fig. 3. Dimensionless velocity profiles in HABON G and ET/NA solutions.
Obr. 3. Bezrozměrný rychlostní profil pro roztoky HABON G a ET/NA.



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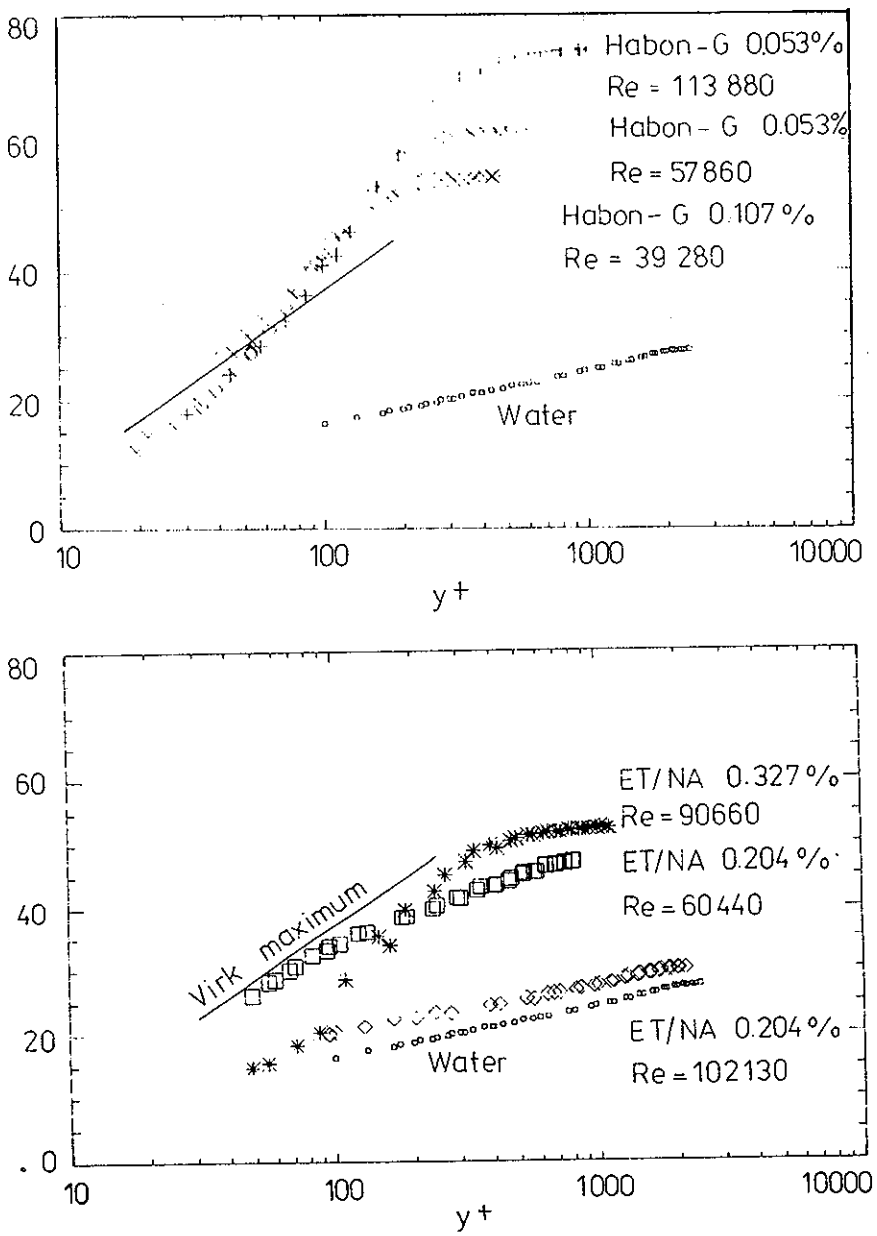


Fig. 4. Velocity profiles in $u^+$ and $y^+$ coordinates.
Obr. 4. Rychlostní profily v $u^+$ a $y^+$ souřadnicích.



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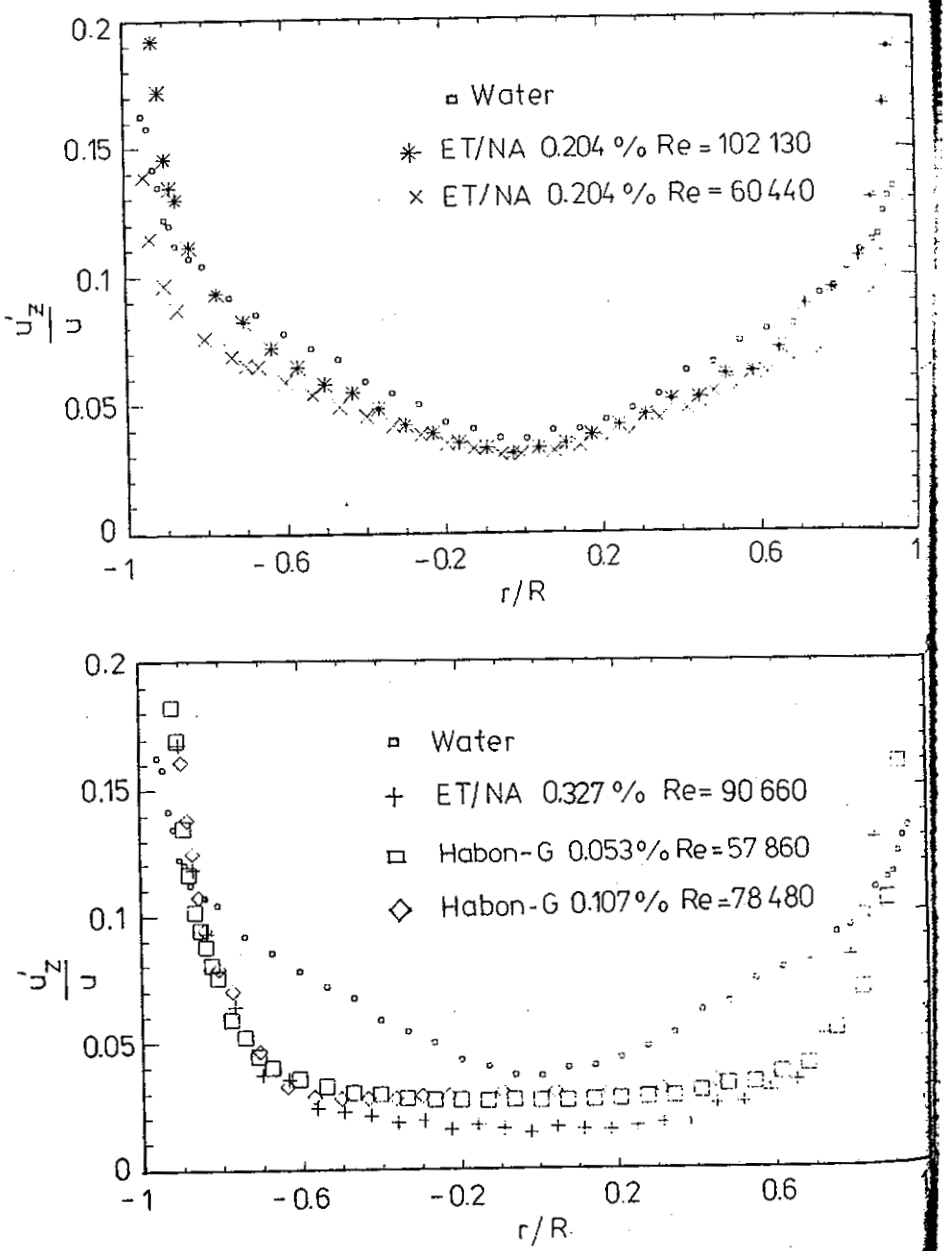


Fig. 5. Intensities of turbulence of the longitudinal velocity component.
Obr. 5. Intenzity turbulence podélné složky rychlosti.



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The S-shape profiles with slopes steeper than Eq.(3) shown in Fig. 4 resemble the other velocity profiles in the literature for systems exhibiting large drag reduction (for example, the data of Bewersdorf and Ohlendorf [1] for tetradecyl-trimethyl-ammonium salicylate).

The profiles of velocity fluctuations in the z direction were also measured for water and for both surfactants and the results are shown in Fig. 5. The ratio of longitudinal fluctuations to the point velocity is generally smaller for the surfactants in most of the cross section. They exceed the values for water in the wall region. The shape of the fluctuation profiles of low concentrated ET/NA solution is similar to the water profile (Fig. 5a). The solutions of ET/NA (0.327%) and Habon G (0.053% and 0.107%) have a tendency to reduce the fluctuations in the central region, where the flat velocity profiles were observed (Fig. 5b).

## Acknowledgements

This research was supported in part under Grant 12.974E, Program in Science and Technology Cooperation Office of Science Advisor, U.S. Agency for International Development. Financial support by the Grant Agency of Czech Republic No. 103/93/1050 is also gratefully acknowledged.

*List of symbols*

| | |
|---|---|
| $i$ | – pressure drop [M L$^{-2}$ T$^{-2}$], |
| $u$ | – local velocity [L T$^{-1}$], |
| $\overline{u}$ | – average velocity [L T$^{-1}$], |
| $u_{max}$ | – maximum velocity [L T$^{-1}$], |
| $u_*$ | – friction velocity [L T$^{-1}$], |
| $y$ | – distance from the wall [L], |
| $u^+ = \frac{u}{u_*}$ | – dimensionless velocity, |
| $y^+ = \frac{y \cdot u_*}{\nu}$ | – dimensionless distance. |

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# MĚŘENÍ TURBULENTNÍCH CHARAKTERISTIK MICELÁRNÍCH LÁTEK SNIŽUJÍCÍCH TŘENÍ METODOU LDA

Zdeněk Chára, Jiří Myška, Miroslav Severa, Jacques L. Zakin

Obsahem předložené práce je experimentální výzkum turbulentních charakteri micelárních aditiv, které mohou být použity např. v rozvodech ústředního vytápění. tyto pokusy byly vybrány dvě průmyslově vyráběná aditiva HABON G a ETHOQU T/13. Na obr. 2 jsou vykresleny tlakové ztráty při proudění těchto aditiv. Jak je patrn obr. 2, účinnost obou micelárních aditiv je poměrně značná. Byly měřeny bodové rychl v kruhovém horizontálním potrubí o vnitřním průměru 39,4 mm. Z provedených měř vyplývá, že při proudění obou aditiv vzniká ve střední části profilu téměř pístový s plochým rychlostním profilem. Výsledky měření intenzity turbulence podélné slo rychlosti ukazují, že se v oblasti plochého rychlostního profilu objevuje prakticky k stantní průběh intenzity turbulence podélné složky rychlosti, který je značně menší ne čisté vodě.

*Seznam použitých symbolů*

| | |
|---|---|
| $i$ | – tlakový spád [M L$^{-2}$ T$^{-2}$], |
| $u$ | – bodová rýchlosť [L T$^{-1}$], |
| $\overline{u}$ | – střední průřezová rychlost [L T$^{-1}$], |
| $u_{max}$ | – maximální rychlost [L T$^{-1}$], |
| $u_*$ | – třecí rychlost [L T$^{-1}$], |
| $y$ | – vzdálenost od stěny [L], |
| $u^+ = \frac{u}{u_*}$ | – bezrozměrná rychlost, |
| $y^+ = \frac{y \cdot u_*}{\nu}$ | – bezrozměrná vzdálenost. |



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# SURFACTANT DRAG REDUCERS FOR REDUCING ENERGY DEMAND IN DISTRICT HEATING SYSTEMS

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ABSTRACT. *The use of drag reducing surfactants in district heating systems is a very promising* method for saving of energy. With this in mind test were carried out in different pipe diameters with different surfactants under different concentration and temperature conditions. Results showed the influence of pipe diameter and of temperature on drag reduction values.

## 1. INTRODUCTION

*District heating world-wide gains in significance in regard to supply the space heating demand.* It is able to promote a primary energy saving, environmentally acceptable and low-priced energy supply. Special advantages of district heating are waste heat utilisation and energy supply by combined heat and power plants. High investment costs of the heat transport and distribution network with relative small annual utilisation times are disadvantages of district heating. Thus all efforts to improve the economy of heat supply is of importance. One of the promising possibilities is using drag reducing additives in district heating systems.

In many closed loops heating systems, such as hot water district heating systems, a drag reduction additive could reduce pumping costs, or increase the potential distribution distance without adding more pumps. For these applications ideally the drag reduction surfactant additive would provide good drag reduction over most of the closed-loop system. The drag reduction additive must be able to withstand the continuous shearing caused by recirculation [1].

The additive is a specially developed surfactant that shows a good stability against temperatures and shear stress. These properties are important for the use in district heating systems. When the substance is added to water, the molecules will assemble in positively charged so-called micelles. The micelles are assumed to form a cubic superlattice and when the water flows, the micelles are assumed to orient in the direction of the flow. Hereby a stratification occurs, which according to the theory - reduce the turbulence. Rod like micelles or possible networks are very effective in reducing turbulent friction up to a certain shear stress, whereupon the micellar bonds or the networks are broken and the fluid becomes Newtonian, with no friction reduction whatsoever. If the stress is relaxed, the bonds reform and the friction reducing ability reappears. This is in contrast to polymer solutions where, if the polymer molecule is degraded or broken, the fluid loses its drag reducing ability permanently [2].

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## 2. EXPERIMENTAL

### A. surfactant additives

Measurements were made using the four quaternary ammonium salts listed in Table 1 with their trade names, suppliers and chemical compositions.

Table 1. List of tested surfactants

| ADDITIVE | SUPPLIER | CHEMICAL FORMULA |
|---|---|---|
| Habon G | Hoechst GmbH (FRG) | $[C_{16} H_{33} N (CH_3)_2 (C_2 H_4 OH)]^+$ |
| Ethoquad T/13 | Akzo Chemicals (USA) | $R^+N (CH_2 CH_2 OH)_3 CH CO_2^-$ |
| Arquad 16-50 | Akzo Chemicals (USA) | $[C_{16} H_{33} N (CH_3)_3]^+Cl^-$ |
| Kemamine Q2983C | Humbo Chemical.Div., Witco (USA) | $[C_{22} H_{43} N (CH_3)_3]^+Cl^-$ |

Except of Habon G surfactant solutions were prepared by adding sodium salicylate (or other anion) solutions to thoroughly mixed quaternary ammonium salt dispersions or direct dissolving of salt in water. Surfactant mixtures were stored 24 hours before testing.

### B. Pipe flow test loop

The test sections for the drag reduction measurements with Habon G and Ethoquad T/13 in Prague's laboratory consist of straight stainless steel tubes of different diameter (4, 6, 10 and 20 mm) each of lengths 5 m. Other parts of the recirculation test pipe loop were constructed from PE smooth tubes of D = 20 mm and all valves are of the same size. The entrance length to the first pressure tap was at least 150 times the diameter of each tube used in order to insure that the turbulent flow was fully developed. The pressure measurements were made using Hottinger Baldwin differential pressure transducers with distance between pressure taps 1 m. Each pressure tap was equipped with two independent pressure transducers. The flow rate of the test fluid was measured by a Krohne magnetic - inductive flowmeter . Its accuracy was ± 0.1 % for flow rates above 10 % of full flow. All data, including temperatures, were collected into a Multipoint Measuring Instrument UPM 60 ( Hottinger Baldwin). The temperature of the test fluids could be controlled from 10 to 60° C to ± 0.5 ° C.

All of the friction factor versus Reynolds number and temperature data was obtained by using a computer programme in connection with UPM 60. It automatically collected and averaged 50 pressure readings and flow rate readings for one experimental point at a rate of about 1 reading/0.2s. The friction factor, $\lambda$, as a function of the Reynolds number was then calculated for these data from the properties of water and the equipment parameters.

The pumping system consisted of a centrifugal pump with a maximum capacity of 78 l/min and H = 70 m.

Control tests were run on Prague's tap water (used for all tests) from 20 to 60° C and compared with the Prandtl-von Kármán friction factor relation. Results for water at all temperatures showed excellent agreement with values predicted from the calculations. All surfactant additive experiments were carried out at three temperatures - 20, 40 and 60° C. Duration of each concentration test was 15 hours.

For testing other surfactants - Kemamine Q2983C and Arquad 16-50 - was used Ohio State University test loop: D = 6.2 mm, distance between pressure taps 0.5 m, stainless steel pipe. The pumping system consisted from centrifugal pump with bypass and a flow control valve. The flowrate was monitored by Omega FPM 5200 paddle-wheel sensor with accuracy ± 1 % of the maximum flowrate. The pressure drop measurements were made with Validyne pressure inductive transducers. The accuracy of the transducers was ± 1 % of full scale. Entrance length was 300 D.

## 3. RESULTS AND DISCUSSIONS

One of the most interesting issues that has surfaced during studies of drag reduction by polymers is the "diameter effect". This refers to the fact that for a given fluid and Reynolds number,

the friction (except in the maximum drag reduction asymptote regime [3] is a function of the diameter of the pipe, i.e. for a given polymer and concentration the drag reduction is greater in small pipes. This is, of course, not the case with Newtonian fluids for which the pipe diameter is already taken fully into account in the Reynolds number.

Drag reduction data for 0.05 % Habon G and Ethoquad T/13 solutions are shown in Figs. 1, 2 and 4 for four different pipe diameters at two different temperatures. Data from [4] and [5] in a 39.4 mm pipe are also shown. The friction factor in some cases falls below the maximum drag reduction asymptote found for polymer solutions [3]. At high values of the wall shear stress, the solution becomes ineffective for drag reduction and behaves like the solvent. The influence of pipe diameter is strong, i.e. with increased pipe size, drag reduction increases as does the range of Reynolds number in which the surfactant is effective. This is contrary to results for polymer additives where drag reduction increased with decrease in pipe size[6]. Similar results were observed with 0.1 % Habon G, too.

The date for all experiments showed strong temperature influence, i.e. as the temperature increased the drag reduction also increased in the range of studied temperatures. Fig.3 show the percentage drag reduction at various temperatures for water with mixture of two surfactants: Kemamine Q2938C and Arquad 16-50 with salicylate salt (concentration ratio 4mM : 1mM : 12.5 mM).

For both surfactants at all concentrations that were studied, the results fall into two distinct groupings; one of the smaller size pipes: 4 and 6 mm and the other, the larger pipes 10 and 20 mm (it is also possible to include the results from the large pipe D = 39.4 mm in this group [6, 7). In the smaller pipes the critical wall shear stress was reached at low Reynolds number and sharp changes in drag reducing effectiveness in a narrow range of Reynolds number were not observed. But in the cases of the 10 and 20 mm pipe sizes it was possible to see typical surfactant drag reduction additive behaviour is seen in which the stress causes breakdown of the network structure of the rod-shaped micelles when the shear stress exceeds a critical value.

It is not clear why drag reducing data fall into two groupings. One possible explanation is related to the differences in the shapes of the velocity profiles with surfactant and without surfactant [4]. Velocity profiles in drag reducing micellar solution systems are quite flat in the central region [5]. In the smaller 4 and 6 mm tubes, the size of the micelles may be large relative to the distance from the wall at which the expected steep portion of the profile should lie, thus interfering with the development of the profile.

Buildings heating and cooling systems are usually quite complicated in configuration, typically involving many fittings, control valves, pumps, and heat exchangers . One expects that these components could perhaps interfere with the viscoelastic drag-reducing fluid, thereby affecting its overall drag and heat transfer characteristics. On the other hand, the surfactant additives could also influence the characteristics and performance of these components. Accordingly, they have studied the effect of drag-reducing surfactant solutions on the performance of centrifugal pumps, and found in some cases' improvements in pump efficiency and cavitation conditions. On the other hand, pumps and valves were shown to have very little localised effect on the drag reduction effectiveness of the fluid. Therefore present experimental investigation was mainly focused on finding of influence of chemical structure, diameter and temperature effect on overall drag reduction.

## 4. CONCLUSIONS

The application of cationic surfactants causes a reduction of pressure loss and also a decrease of heat transfer in turbulent flow and from the experiments it is possible to conclude:

1. For the surfactants, the influence of pipe diameter on drag reduction is opposite to that of polymers, i.e. with increasing diameter drag reduction increased for the surfactants.
2. In the range of experimental temperatures ($t_{max}$ = 120$^{o}$ C), the maximum drag reduction increased with increase of temperature.
3. During experiments in a closed loop pipeline system no mechanical or ageing degradation effects were observed as are found with polymer additives.
4. Friction factors lower than those predicted by the maximum drag reduction asymptote.

Tab.1 - Drag reduction results (%) for different pipe diameters and temperatures
N - no experimental data; Habon G - concentration 0.05 %;
data for D = 39.4 mm from [4]

| | t (°C) | | t (°C) | | t (°C) | | t (°C) | |
|---|---|---|---|---|---|---|---|---|
| | 20 | 60 | 20 | 60 | 20 | 60 | 20 | 60 |
| D (mm) | Re = 10000 | | Re = 20000 | | Re = 50000 | | Re = 60000 | |
| 4 | 5 | 43 | 0 | 5 | 0 | 0 | N | N |
| 6 | 37 | 57 | 14 | 19 | 7 | 0 | N | N |
| 10 | 63 | 74 | 14 | 79 | N | 79 | N | 75 |
| 20 | 71 | 73 | 73 | 81 | N | 80 | N | 80 |
| 39.4 | N | N | N | N | N | N | 81 | N |

Because of this the following options could be realised:

a. in existing plants

- reduction of pressure loss at the same flow-rate, resp. an increase of flowrate at the same pressure difference
- an increase of throughput and by this an increase of heat load at the same power an temperature difference

b. in new designed plants

- smaller tube diameters or reduced pressure losses and with this reduction of pump power
- heat supply by large power stations quite far away from the consumers.

The reduction of pressure loss in transportation and distribution pipes has essential advantages for district heating. The deterioration of heat transfer "water/tube wall" has nearly no advantages, because in these tubes the outer heat transfer resistance is controlling the heat loss. An enlargement of heat transfer area of heat exchangers might be required.

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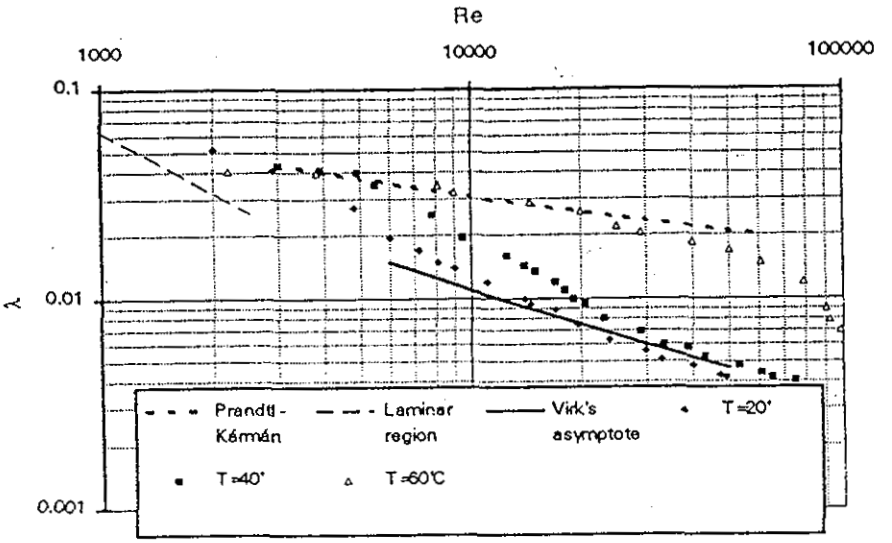


Fig.1- Relationship of Reynolds number and friction factor for solution of 0.2% Ethoquad T/13 with NaSl in 10 mm pipe.

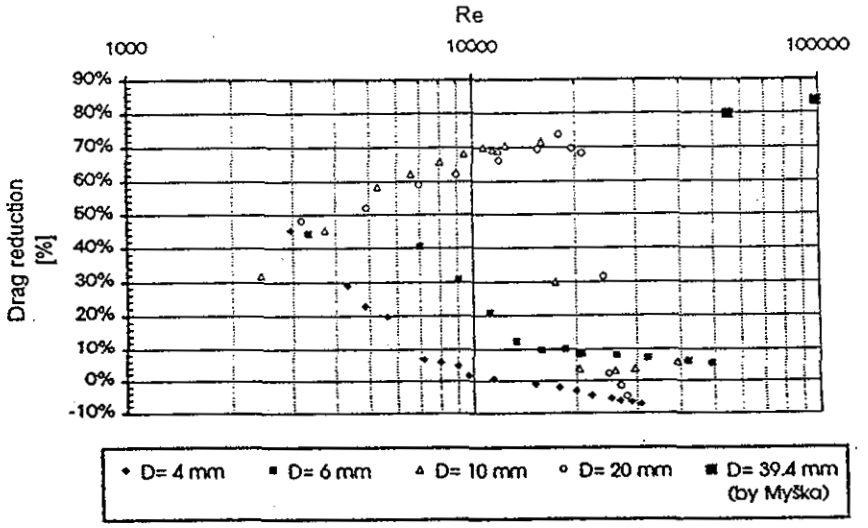


Fig. 2 Changes of drag reduction in different sizes of pipes under isothermal condition, t=20°C

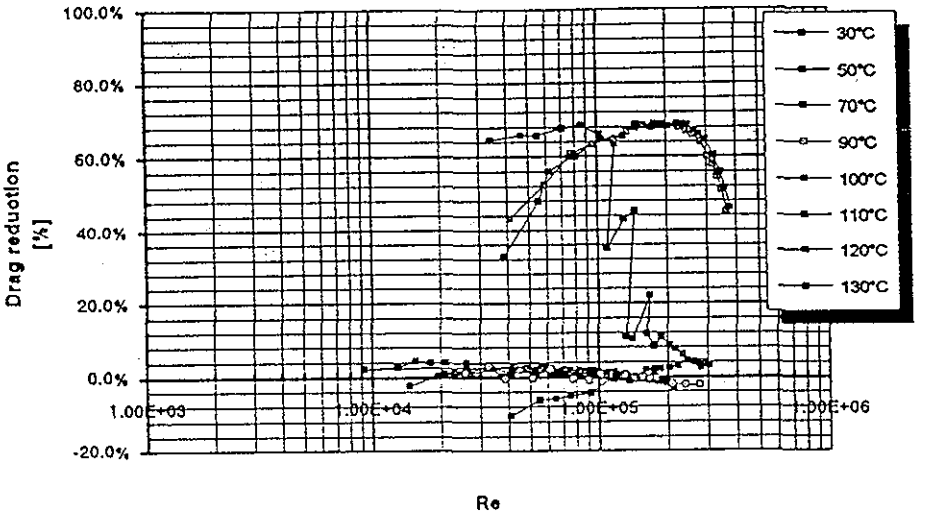


Fig. 3- Influence of temperature on drag reduction of mixed surfactants Kemamine Q2983C + Arquad 16-50 + 5 Cl-Salycilate (concentration ratio: 4mM/1mM/12.5mM), D=6.2 mm

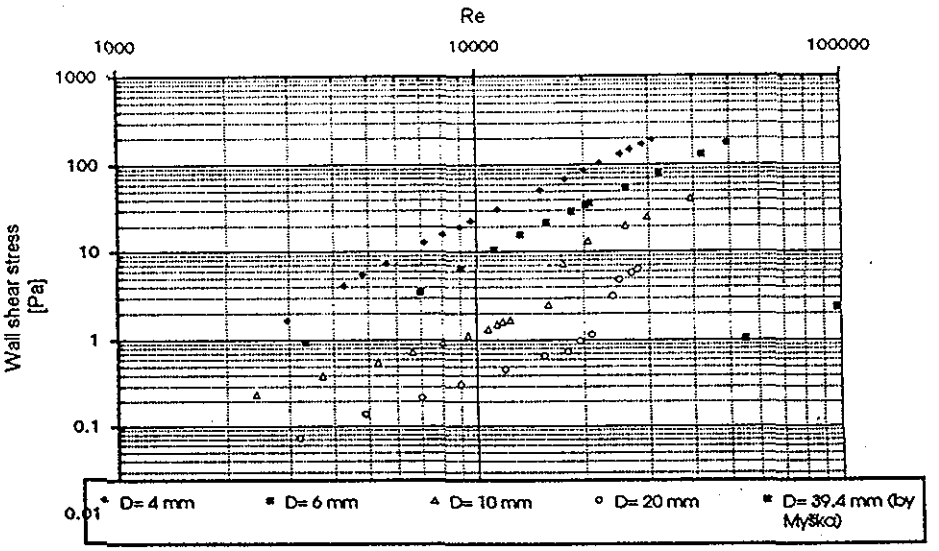


Fig.4- Influence of pipe diameter on wall shear stress, isothermal flow conditions, concentration of Habon G 0.05%, t=20°C



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# RELATIONSHIP OF FLOW BIREFRINGENCE AND NORMAL STRESSES OF CATIONIC SURFACTANT SYSTEMS TO THEIR TURBULENT FRICTION REDUCTION CHARACTERISTICS

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## 1. INTRODUCTION

The use of district heating and cooling systems (DHC) is being encouraged in many countries in order to make more effective use of energy resources and to reduce environmental pollution by combustion products. The addition of drag reducers to these systems could reduce pumping energy costs, reduce capital investments or increase capacity and further reduce environmental problems.

Certain surfactants consisting of quaternary ammonium salts with substituted benzoates or naphthoates as counterions form rod-like micelles in aqueous solutions, exhibit viscoelastic behavior and are effective as drag reduction additives. They regain their drag reduction effectiveness after mechanical degradation at high shear stress or if their temperature limits are exceeded.

In this paper, flow birefringence and normal stress measurements on three cationic surfactant systems are reported and compared with drag reduction measurements.

## 2. EXPERIMENTAL

### 2.1 Cationic Surfactants and Counterions

The three cationic surfactants studied are Arquad 16-50 ($C_{16}H_{33}N(CH_3)_3Cl$), Ethoquad O-12 (Oleyl-$N(CH_3)(C_2H_4OH)_2Cl$) and Ethoquad T/13-50 (Tallow-$N(C_2H_4OH)_3Ac$). All three are commercial products of Akzo Chemical, U.S.A. The counterion, the sodium salt of 2-hydroxy benzoate (NaSal), is a product of MCB Manufacturing Chemicals, Inc. (GR purity grade).

All three solutions were prepared with distilled water at concentrations of 5mM surfactant and 12.5mM NaSal.

### 2.2 Drag Reduction Measurements

Drag Reduction experiments at temperatures from 2 °C to 130 °C were carried out in a recirculation system containing a 122 cm long, 0.617 cm diameter stainless steel test section. Entrance and exit corrections were applied [Chou, 1991].

### 2.3 Flow Birefringence Measurements

Polarized light passing through an isotropic material experiences a single, scalar refractive index. In a birefringent material, this light experiences a tensorial refractive index. Flow birefringence is due to anisotropy in the refractive index. Certain surfactant systems, because of the existence of rod-like micelles, show birefringent behavior.

A phase-modulated flow birefringence apparatus was employed [Smith, 1992]. A beam of monochromatic, partially polarized red light from a He-Ne laser was linearly polarized when passed through an incident polarizer. It then passed through a photoelastic modulator which induced a time varying retardance in the phase of the light. The beam then traveled through the solution in a Couette cell with outer cylinder rotation and a one millimeter gap size. Shear rates could be varied from 0 to 700 1/s. The cell was jacketed for temperature control. The light then passed through a second polarizer and into a PIN photodiode detector connected to computer data acquisition.

The flow birefringence and the extinction angle of the solutions were simultaneously determined.

### 2.4 First Normal Stress Difference Measurements

The first normal stress difference, N1, was measured in a Haake CV 20N rheometer with a



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cone-plate system (range: 0-600 1/s) over temperature range of 20 °C to 80 °C. The cone is of diameter 41.7mm and the angle is 2°. The measurements were controlled by a computer using Haake software.

The solutions were presheared before the measurements of N1 by shearing in a Couette flow cell for 2 hours at shear rate of 1170 1/s.

## 3. RESULTS AND DISCUSSION

### 3.1 Correlation of Three Measurements

All three systems are good drag reducers up to 70 °C (Table 1). The temperature ranges for flow birefringence are in good agreement with those of drag reduction except for the Ethoquad T/13-50 system. The first normal stress difference upper temperature limits for the Ethoquad O-12 and T/13-50 systems are lower than those for drag reduction.

**Table 1. Comparison of Temperature Range for Three Measurements**

| System (with NaSal) | Drag Reduction (°C) | Birefringence (°C) | Normal Stress (°C) |
|---|---|---|---|
| Arquad 16-50 | 10-70 | 30*-70 | 20*-70 |
| Ethoquad O-12 | 2*-70 | 30*-70 | 20*-50 |
| Ethoquad T/13-50 | 2*-80 | 20*-40 | 20*-60 |

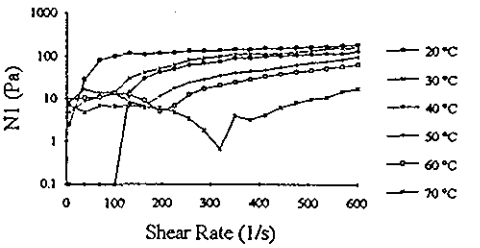
* The lowest temperature measured.

The existence of rod-like micelles in the solution is believed to be essential for the existence of drag reduction, flow birefringence and viscoelasticity. The discrepancies in the upper temperature limits suggest that these properties depend on the size of the rod-like micelles to different extents.

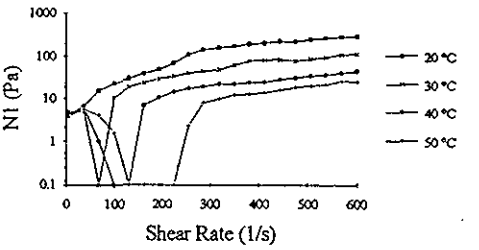
### 3.2 First Normal Stress Difference

The first normal stress difference values, N1, of all systems rise above $10^2$ Pa at 20 °C (Fig. 1). The normal stresses decrease with increasing temperature for all systems with the exception of the Ethoquad T/13-50 system at 30 °C in the range of shear rates smaller than 330 1/s, whose values are lower than those at 40 °C. At a certain shear rate, the normal stress decreases to a minimum and then increases with shear rate. This phenomenon was observed in all three systems at all temperatures except for the Arquad 16-50 and Ethoquad O/12 systems at 20 °C.

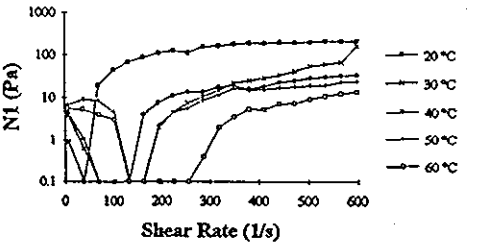
With the exception of the Ethoquad T/13-50 system at 30°C, each system shows an approximately constant critical shear stress above which normal stress values increase monotonically and which is apparently not dependent on temperature (Table 2). Thus, we may conclude that at a certain shear stress, the structure of micelles in the solution is changed so that the behavior of the first normal stress difference undergoes an abrupt change.




(1a) Arquad 16-50/NaSal (5mM/12.5mM)




(1b) Ethoquad O-12/NaSal (5mM/12.5mM)




(1c) Ethoquad T/13-50/NaSal (5mM/12.5mM)

**Fig. 1 First Normal Stress Difference Results**

**Table 2. Critical Shear Stress for Normal Stress Minimum**

| System | Temperature (°C) | Shear Rate (1/s) | Shear Stress (Pa) |
|---|---|---|---|
| Arquad 16-50 + NaSal | 30 | 69 | 0.60 |
| | 40 | 100 | 0.68 |
| | 50 | 162 | 0.77 |
| | 60 | 194 | 0.72 |
| | 70 | 319 | 0.54 |
| Ethoquad O-12 + NaSal | 30 | 69 | 0.65 |
| | 40 | 131 | 0.74 |
| | 50 | 225 | 0.86 |
| Ethoquad T/13-50 + NaSal | 20 | 38 | 0.32 |
| | 30 | 162 | 0.66 |
| | 40 | 131 | 0.24 |
| | 50 | 162 | 0.22 |
| | 60 | 256 | 0.26 |

### 3.3 Flow Birefringence

Flow birefringence increases with the shear rate to a saturation value for all systems. Fig. 2 shows results for the Arquad 16-50 system. With increasing temperature, the shear rate required to reach the saturation point is increased. This is consistent with the assumption that micelle length decreases with temperature. The shorter micelles are not as easily oriented as the longer ones, and therefore, require a higher shear rate for alignment. Also this is in agreement with the N1 results, which shows that N1 values decrease with increasing temperature because of the shorter micelles which show reduced viscoelasticity.

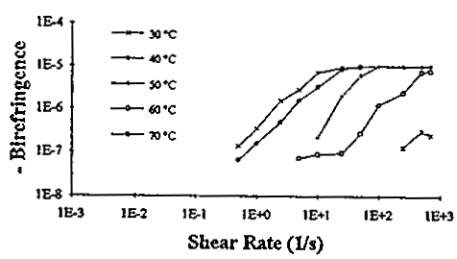



**Fig. 2 Flow Birefringence vs. Shear Rate for Arquad 16-50/NaSal (5mM/12.5mM)**

### 3.4 Stress-Optical Law

The flow birefringence, $\Delta n$, and the orientation angle, $\chi$, can be linearly related to the shear stress and the first normal stress difference through the Stress-Optical Law [Coleman, et al., 1970, Janeschitz-Kriegl, 1983]. This law states that there is a simple proportionality between the refractive index tensor, **n**, and the stress tensor, $\tau$:

$$\mathbf{n} = C \bullet \tau \quad \text{----------} (1)$$

where C is the stress-optical coefficient. In component form, the law states:

$$\Delta n \bullet \sin(2\chi) = 2C\tau_{12} \quad \cdots\cdots\cdots\cdots\cdots\cdots (2)$$

$$\Delta n \bullet \cos(2\chi) = C(\tau_{11} - \tau_{22}) \cdots\cdots\cdots\cdots (3)$$

where $\tau_{12}$ is the shear stress and $(\tau_{11}-\tau_{22})$ is the first normal stress difference.

The validity of this law for polymer systems was checked by many researchers [Coleman, et al., 1970, Janeschitz-Kriegl, 1983, Osaki, et al., 1979]. However, there are no reports testing this law for surfactant systems.

These surfactant systems can not follow Eq. (2), because as shear rate increases, the orientation angle, $\chi$, becomes zero so that the left hand side of Eq. (2) is zero while the shear stress doesn't vanish with increasing shear rate and in fact increases. Fig. 3 shows orientation angle and shear stress for the Arquad 16-50/NaSal system.

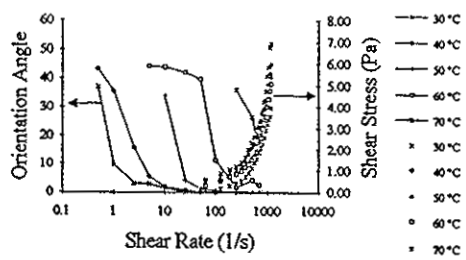



**Fig. 3 Orientation Angle and Shear Stress for Arquad 16-50/NaSal (5mM/12.5mM)**

C can be calculated from Eq. (3) for the three systems. At low shear rate (<300 1/s), C is erratic. However, at shear rates higher than 300 1/s, C changes with the shear rate only to a small extent (Fig. 4).



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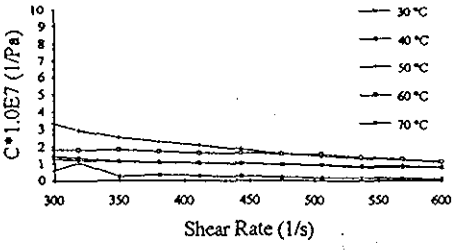
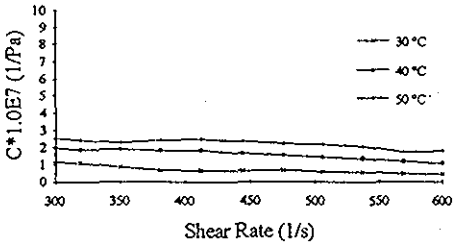
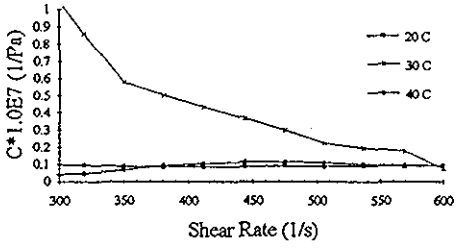



(4a) Arquad 16-50/NaSal (5mM/12.5mM)




(4b) Ethoquad O-12/NaSal (5mM/12.5mM)




(4c) Ethoquad T/13-50/NaSal (5mM/12.5mM)

**Fig. 4 Stress Optical Coef. vs. Shear Rate**

Again, there is an exception for 30 °C data of the Ethoquad T/13-50 system.

The reason for the breakdown of this law for shear stress for these surfactant systems appears to be that the orientation of the rod-like micelles in solution ($\chi$=0) occurs at low shear rate. The near zero value of $\chi$ doesn't affect the first normal stress difference relationship because the cosine function of $\chi$ is involved (see Eq. (3)).

Compared with polymer solutions of similar concentration, surfactant systems have low viscosity (of the order of $10^{-3}$ Pa s) and these micelles are more rod-like in shape. Thus the surfactant micelles can be easily aligned in the flow direction and show low shear stress at the shear rate at which $\chi$ reaches zero.

## 4. CONCLUSIONS

The rough correspondence among drag reduction, flow birefringence and first normal stress difference measurements supports the suggestion that rod-like micelles, which may form networks, are responsible for the existence of these three phenomena. The increase in the shear rate required to reach birefringence saturation and onset of first normal stress difference with increasing temperature is consistent with the assumption that micelle length decreases with temperature as does the disappearance of drag reduction at high temperature. Surfactant systems seem not to be able to follow Eq. (2) due to their low viscosity and the ease which they align under shear. However, in general, the stress optical constant in Eq. (3) shows modest variations with shear rate above 300 1/s.

## 5. ACKNOWLEDGMENT

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# USE OF FRICTION REDUCING ADDITIVES IN DISTRICT HEATING SYSTEM FIELD TEST AT KLADNO - KROCEHLAVY, CZECH REPUBLIC

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## ABSTRACT

The use of drag reducing surfactants in district heating systems is a very promising method for saving energy. With this in mind field tests in a secondary system in Kladno-Krocehlavy were carried out with the drag reducing surfactant, Habon G, under different flow and thermal conditions. Results show the influence of surfactant concentration on drag reduction and heat transfer.

KEY WORDS: Drag reduction, turbulent pipe flow, field test, heating networks, surfactants

# USE OF FRICTION REDUCING ADDITIVES IN DISTRICT HEATING SYSTEM FIELD TEST AT KLADNO - KROCEHLAVY, CZECH REPUBLIC

## INTRODUCTION

As soon as the existence of drag reduction from soluble high polymers and later by surfactants had been established, proposals were made for possible future applications. Over the years this list of possible areas has increased to include: oil well operations, fire fighting, irrigation, sewage and floodwater disposal, hydrotransport of solids, crude oil movement by pipeline, water heating circuits, jet cutting, hydraulic machinery, marine applications and biomedical applications. A recently published paper (1) considers progress in all these application fields, except the biomedical one, and the present article will restrict it's attention to one topic: heating circuits.

Hydrodynamic conditions in heating circuits (primary as well as secondary) are rather different from those in other flow systems usually considered. The salient features are[1]:

- completely closed circulation systems.
- pipe internal diameters can range up to 1.5 m which is several times greater than those in most other applications.
- water recirculation time ranges from minutes to several hours at Reynolds numbers of $10^4$ - $10^6$
- water temperature range of 40 - 130$^o$ C

World-wide use of district heating to supply space heating demands is growing, because of its effectiveness in reducing fuel use, reducing environmental pollutants and its ability to provide a reliable low-cost energy supply. Special advantage can be taken of waste heat and energy from combined heat and power plants. High investment costs of the heat transport and distribution network with relatively small annual utilization times are disadvantages of district heating. Thus efforts to reduce energy use and investment cost are of importance. One of the promising possibilities is to use drag reducing additives in district heating systems.

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In many closed loop heating systems, such as hot water district heating systems, a drag reduction additive can reduce pumping costs, or increase the potential distribution distance without adding more pumps. Ideally, for these applications, the drag reduction additive would provide good drag reduction over most of the closed-loop system. The drag reduction additive must be able to withstand the continuous shearing caused by the pump and by recirculation [2].

While high polymer drag reducing additives are very effective when they are injected into "once-through" petroleum pipelines, they are very sensitive to mechanical shear. One pass through a pump degrades them irreversibly and they are useless downstream. Certain surfactants, on the other hand, provide good drag reduction and they recover immediately after passing through regions of high shear such as in pumps.

The drag reducing cationic surfactant additive used in these experiments, Habon G, is a specially developed surfactant that shows good stability at high temperature and shear stress. When the substance is dissolved in water, the molecules assemble to form large rod-like aggregates called micelles. The micelles are believed to form a cubic superlattice and, under shear, the micelles orient in the direction of the flow. Rod-like micelles or possible networks are very effective in reducing turbulent friction up to a certain shear stress, at which point the micellar bonds or the networks are broken and the fluid becomes Newtonian and loses its friction reducing character. If the stress is reduced, the bonds reform and the friction reducing ability reappears. This is in contrast to polymer solutions where, if the polymer molecule is degraded or broken, the fluid loses its drag reducing ability permanently.

During the past ten years considerable research, development and demonstration of technology for the use of drag reducing additives for district heating and cooling has been going on.

The results reported here are based on an additive developed by Hoechst AG in Germany. The additive is a specially developed surfactant, cetyl dimethyl hydroxyethyl ammonium 3-hydroxy-2-naphthoate (Habon G). This additive or a similar product demonstrated effectiveness in primary heating networks in Denmark and in Germany. The experiment in Denmark was carried out in a 2.8 km long pipe running from the power plant of Herning to the town of Lind, D = 200 mm in the primary heating circuit. A number of measurements at various concentrations of Habon G were made and results showed that the pressure loss for the entire system was reduced by 75 % or more at concentrations greater than 250 ppm. Measurements were made at concentration up to 1000 ppm. The maximum effect, however, appeared at 460 ppm [3].

In Germany the first full scale experiments were carried out with drag reducing surfactants in 1988 in a district heating transport pipe, D = 450 mm, connecting the heating and power station at Fenne and the district heating central station at Volklingen [4]. The transport duct which was 2400 m total length was fed by a pump of nominal 1450 $m^3$/hour with a delivery head of 67 m. A steam heated tube bundle heat exchanger in the heating and power station at Fenne (ca. 30 MW) was used for heat input and a plate heat exchanger (10 MW) within a central station was used to heat the secondary system water. In the whole primary heating system, pressure drop decreases up to 70 % were observed or alternatively, flow rate increases up to 30 %. The surfactant used was Obon (octadecyl trimethyl ammonium 3-hydroxy-2-naphthoate) also produced by Hoechst AG.



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## THE COOPERATIVE INVESTIGATION PROJECT

Despite the experience gained in these tests, questions remain on how drag reduction by surfactants can be implemented on a full scale. One of the most important is how effective will the friction reducing solution be in a secondary system and how will it affect heat transfer in heat exchangers of combined heat and power plants.

Another key issue is the effect of the additives on the environment. The drag reducing additives available from Hoechst, AG have properties very similar to surfactants widely consumed - and disposed of - in modern society. A distinct difference in the application in district heating systems is the fact that the additives are to be used at fairly low concentrations (200 - 1500 ppm) in closed loops only. Only in case of accidents is there a risk of their admittance to the environment, and even in this case the consequences are limited. In general, the risk of leaks from transmission networks to the environment is small. If it should occur, however, the adsorption of additives into the surroundings such as soil, is very high and adsorption is irreversible. Due to toxicity to fish, because of the low surface tension of the surfactant solution, surfactant discharges into surface waters are more critical and precautions to avoid this need to be taken.

The Czech Republic is the most developed country of the former Central and Eastern European countries in terms of central delivery of heat into households by district heating systems. Today there are nearly 100 central stations operating with total heat capacity above 100 MW, i.e. ca. 32 % of the flats and many other buildings are connected by pipeline to such heat stations. Another 18 % of flats and houses have local heating stations.

The total length of primary heating circuits (100 % of them use water as the source of heat) is 8000 km and secondary heating circuits (which also use 100 % hot water with maximum temperatures up to 92.5 $^{o}$C) have a total length of nearly 15 000 km.

With all these above mentioned problems in mind, an international cooperative project "Reducing Energy in District Heating Systems" supported by the United States Agency for International Development, was initiated in August 1991. The collaborators are The Ohio State University, The Czech Technical University, The Institute of Hydrodynamics of the Czech Academy of Sciences and the Czech Research Energy Institute. A field test of a drag reducing surfactant additive in a district heating system is an important part of that program.

## FIELD TEST IN KLADNO - KROCEHLAVY

For the evaluation of the effect of surfactant drag reducing additives in district heating systems an existing district heating system was used in Kladno - Krocehlavy (25 km west of



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Prague). The network there is used for heating 12-story apartment buildings (typical eastern European houses from prefabricated concrete blocks). The source of heat is a nearby power plant which is part of the well known steel works - Poldi Kladno.

The primary pipeline system delivers hot water to heat exchange stations which are situated in the middle of groups of houses. These stations are connected by secondary systems to blocks of buildings. One of them, consisting of five apartment houses and other smaller buildings, was used for the field test with surfactant additive. A schematic representation of this secondary network is shown in Fig. 1 which includes detailed information about the whole system. Three centrifugal pumps were used to circulate water in the system.

The main parameters of the secondary network are: volume of main distribution pipelines 8.03 $m^3$, volume of 3 tube-in-tube heat exchangers - 1.68 $m^3$, volume of expansion vessel - 2 $m^3$, volume of pipelines in houses and flats - 8.19 $m^3$, volume of radiators in flats (lower floors cast iron and higher floors steel) - 19.78 $m^3$. The total volume was 39.68 $m^3$. Leakage averaged 50 l per day which was replenished continuously. Total capability of the secondary network to supply heat was 2154.9 kW. Design capacity is based on outside temperature of -10 $^oC$, and flow rate of 60 $m^3$/hour. Diameters of pipelines in the system varied from 200 to 10 mm. Water in the secondary system was treated thermally and hardness ranged from 0 - 30 ppm and pH = 8.

At the heating station surfactant was injected by a manually operated piston pump. The station has 3 ten year old tube-in-tube heat exchangers of Czech production with total heat transfer area in each of 40 $m^2$. For operation without additives hot water was pumped by 3 centrifugal pumps of Czech manufacture SIGMA-150-NTC-175-29-LB-00 with parameters: Q = 1620 l/min, Y = 58.8 J/kg, P = 3 kW, n = 1450 rev/min (Fig. 2).

While the previous field tests in Denmark and Germany were on primary systems, this field test program was carried out in a secondary heating network and focused on the following problems:

- investigate the effectiveness of drag reducing surfactants in secondary heating networks.
- evaluate the influence of the secondary system with its many singularities and many changes in flow conditions, in contrast to primary systems or laboratory setups with only one pipe diameter, on drag reduction efficiency.
- determine the optimum concentration of surfactant to achieve the highest drag reduction in the system.
- determine the influence of surfactant additives on heat transfer in exchangers under full load conditions.
- carry out an economic assessment of the project. This assesment is not yet complete.

Tests were carried out with varying concentrations of Habon G. The following test data were obtained:

a. pressure difference in secondary pipeline system, i.e. including radiators in flats, by differential pressure transducer: Rosemount, Switzerland.

b. average volume flow rate by ultrasonic flow meter: UNIFLOW, Comtratron Co, U.S.A.



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c. actual surfactant concentration by analytical method recommended by Hoechst, AG as well as theoretical concentration from direct calculation from total volume of the secondary system (without taking leakage into account).

d. total pump electric power input.

e. temperatures at different locations by resistance electric thermocouples: Pt100.

All data were collected and computerized on PC AT 386.

## FIELD TEST PROCEDURE, RESULTS AND DISCUSSION

In pipe heating systems (primary and secondary) in which the lengths, diameters, surface roughness and many other friction losses in singularities of the network are fixed, the important variables are the pressure gradient ($h_Z/l$) and the discharge rate (Q). Heating and cooling systems in buildings are usually quite complicated in configuration, typically involving many fittings, control valves, pumps, and heat exchangers. These components could interfere with the viscoelastic drag reducing fluid, thereby affecting its drag and heat transfer characteristics. On the other hand, the surfactant additives could influence the characteristics and performance of these components. Studies of the effect of drag reducing surfactant solutions on the performance of centrifugal pumps have, in some cases, shown improvements in pump efficiency and cavitation characteristics. On the other hand, pumps and valves were shown to have little localized effect on the drag reduction effectiveness of the fluid. Therefore the present experimental investigation was mainly focused on studying the influence of surfactant concentration and temperature effects on overall drag reduction.

The test program on the secondary system was started after installation of equipment in late 1992. Preliminary experiments with water began in January and February 1993 and again in November 1993 under different outside temperatures. Results of laboratory investigations showed that the concentration giving the highest drag reduction in different pipe sizes was 400 - 500 ppm.

To better understand the influence of this complicated secondary system on overall drag reduction, it was decided to increase the concentration incrementally from a theoretical 100 ppm (based on the total volume of the system) to 1000 ppm. The first dosing was done on December 1, 1993 and the last on February 24, 1994. During this period the concentration was increased 10 times. Fig. 3 shows the great difference between the actual and the theoretical concentration (in the latter case no leakage or other loss of water from the system was considered), which was caused by adsorption of surfactant on the walls. The initial difference was large with only 7 ppm in the system despite dosing at a theoretical level of 106 ppm. After the last dosing, the theoretical concentration was 1378 ppm and the actual concentration was 623 ppm. Only a minor part of this difference can be attributed to leakage.

Soon after the first dosing, drag reduction was observed (dosing time was 10 min., but it took about an hour for the additive to be dispersed throughout the system), i.e. pressure

dropped and flow rate increased (see Fig. 4). The duration of the effect was short due to the low concentration and the adsorption. When dosing of Habon G was repeated even greater drag reduction was achieved and the effect lasted longer. These first results were very encouraging because they showed that drag reduction by surfactants like Habon G could be effective in a secondary heating network and not only in straight pipes with few singularities, i.e. in primary systems. These first experiments also showed another interesting thing - almost no change in the inlet and outlet temperatures in the heat exchangers (Fig. 4).

Normal operation of the system used three centrifugal pumps which provided an adequate supply of hot water to the radiators. When concentration reached ca. 400 ppm, total reduction of specific pumping energy reached 30 % which was equivalent to the power provided by one of the pumps. At that point the third pump was switched off and the whole secondary system operated with only two pumps without any difficulties in spite of very cold weather during two weeks in February 1994. Fig.5 shows details of the results including changes in the characteristics of centrifugal pumps and specific energy demand of the secondary heating system with different Habon G concentrations. The maximum decrease in specific energy, 40.8 %, was achieved at a concentration of 450 ppm of Habon G. When actual concentration was increased above 372 ppm, no significant increase in drag reduction was observed. Thus, the optimum actual concentration for such a system is 400 - 450 ppm. At this concentration of Habon G, total drag reduction was very stable for several weeks. The last measurements in the system were made on March 18, 1994. Additional measurements will be made at the beginning of the new heating season in October 1994.

Reduction of heat transfer is a general problem in heat exchangers using drag reducing additives (8). During experiments with Habon G in the secondary heating system in Kladno-Krocehlavy, the effect of the surfactant on the overall heat transfer coefficient was small, below 10 % reduction. Fig. 6 shows changes in the overall heat transfer coefficient, U, for different flow rates in the primary system for water and for a 200 ppm solution of Habon G. Average values of flow rate and U were calculated from experimental data (see Appendix). One explanation for the nearly constant overall heat transfer coefficient may be that the walls of the exchangers were at very high temperatures exceeding the critical upper temperature limit for drag reduction of Habon G. In this temperature region the additive solution characteristics revert to those of water. An alternate explanation is that significant scale had built up in the exchanger, so overall resistance was already high. Table 1 shows a comparison of U values at similar secondary system flowrates for water and for a 200 ppm Habon G solution.

Table 1: Influence of Habon G on heat transfer coefficient, U, in secondary network

| | | |
|---|---|---|
| Water | $\overline{Q}_{v_2} = 73.6\ m^3/hour$ | $\overline{U} = 266\ W/m^2/K$ |
| Habon G - 200 ppm | $\overline{Q}_{v_2} = 74.3\ m^3/hour$ | $\overline{U} = 258.7\ W/m^2/K$ |

Performance of the two centrifugal pumps in series was almost identical for water and for surfactant solutions, as shown by the specific energy characteristics of the pumps (Fig. 5) The specific energy requirements of the secondary heating circuit were reduced by the

presence of surfactant by 30 - 40 % when compared to water. This reduction in the secondary circuit specific energy characteristics allows the equilibrium working point of the system to move from flow rate 58 $m^3/h$ for water (point A in Fig. 5) to 70 $m^3/h$ for surfactant solution (point B in Fig. 5), with almost no increase in the power input to the two pumps. If water is used in the secondary system, a flow rate of 70 $m^3/h$ requires all three pumps to be in operation (point C in Fig. 5).

## CONCLUSIONS

Field tests in a secondary heating network demonstrated that:

1. When drag reduction in the secondary system reached 30 %, one of three pumps could be switched off and the whole secondary network operated with two pumps without difficulties.

2. The maximum decrease of specific pumping energy reached 40.8 % at about 450 ppm of Habon G, a surfactant drag reducing additive.

3. In secondary heating networks, drag reduction does not reach the high values observed in primary networks. The difference is caused by the greater number of singularities which causes friction losses which are not reduced by the additive. Also in some parts of the system surfactant solutions may be "ineffective" because the critical shear stress is exceeded.

4. For maximum drag reduction, the optimum concentration of Habon G in solution was 400 - 450 ppm. This is consistent with laboratory experiments in smaller tubes as well as with previous field tests in primary systems.

5. The secondary heating system had a large surfactant adsorption capability requiring introduction into the system of much more surfactant than was actually present in the circulating fluid.

6. No significant effect of the presence of the surfactant drag reducing additive on overall heat transfer coefficient was observed. This may be the result of very hot tube walls in the heat exchanger or of using older heat exchangers which may have built up scale on the metal surfaces which already provide considerable resistance to heat transfer.

Thus, surfactant drag reducing additives provide excellent potential for energy savings. Along with the savings, large benefits to the environment can be obtained. This is due to the reduced demand for pumping power (use of fossil fuels) and extended possibilities for use of surplus heat. On the other hand, potential risks to the environment in case of accidents have to be taken into consideration.



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APPENDIX

The following equations were used for the calculations:

Primary heating network

$$[Q = Q_{v1}\, \rho\, c_p\, (t_1 - t_2)]_1$$

Secondary heating network

$$[Q = Q_{v2}\, \rho\, c_p\, (t_3 - t_4)]_2$$

where

| | |
|---|---|
| $t_1$ | temperature of water in inlet into the heat exchanger: primary network pipeline ($^{o}$ C) |
| $t_2$ | temperature of water in outlet from the heat exchanger: primary network pipeline ($^{o}$C) |
| $t_3$ | temperature of water in outlet from heat exchanger: secondary network pipeline ($^{o}$C) |
| $t_4$ | temperature of water in inlet into heat exchanger: secondary network pipeline ($^{o}$C) |
| $\rho$ | specific mass |
| $Q_{v1}$ | calculated volume flow rate in primary heating network ($m^3$/hour) from heat balance |
| $Q_{v2}$ | measured volume flow rate in secondary network ($m^3$/hour) |
| Q | amount of heat transferred (kW) |
| | $Q = U\,S\,(\Delta T_{LM})$ |
| | $\Delta T_{LM}$ - logarithm mean temperature difference |
| | $c_p$ - specific heat of test fluid |
| | where $\Delta T_{LM} = \dfrac{(t_1 - t_3) - (t_2 - t_4)}{\ln \dfrac{t_1 - t_3}{t_2 - t_4}}$ |
| U | overall heat transfer coefficient ($kW/m^2$) |
| S | area of heat transfer in heat exchangers ($m^2$) |

Note: for all calculations characteristic values for surfactant solutions were taken as those for water at the same temperature.

Fig. 1: General scheme of the secondary heating network used for field test in Kladno-Krocehlavy at heat exchange station No. PS 67

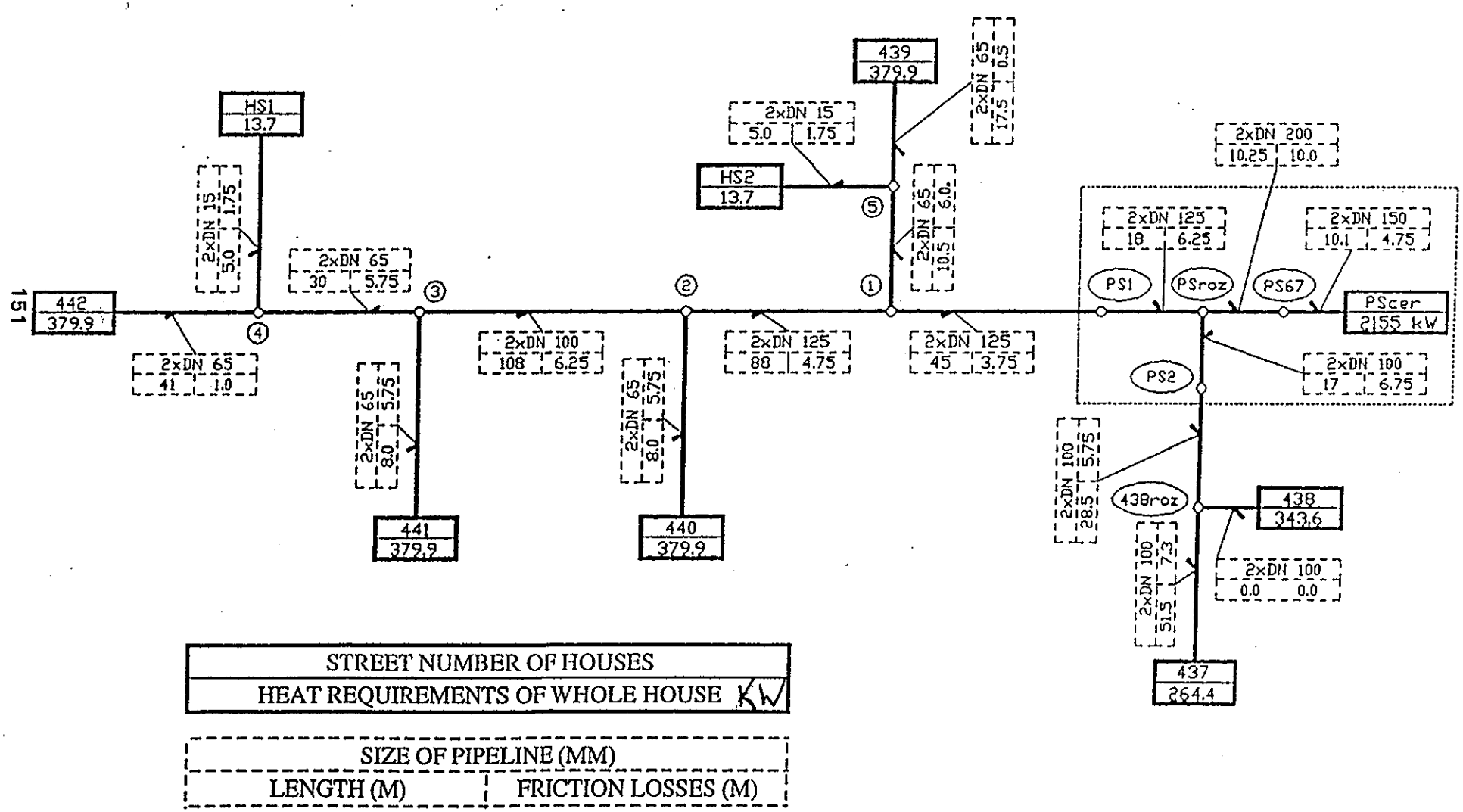


Fig. 2: Schematic diagram of the heat exchanger station and instruments used in Kladno-Krocehlavy; $t_2$ - temperature of outlet water from heat exchanger: primary network pipeline (°C); $t_3$ - temperature of outlet water from heat exchanger: secondary network pipeline (°C); $t_1$ - temperature of inlet water into the heat exchanger: primary network pipeline (°C); $t_4$ - temperature of inlet water into heat exchanger: secondary network pipeline (°C); tz - outside temperature of air (°C); Qv - volume flow rete of water in secondary network ($m^3/h$); Δp - differential pressure transducers (Pa).

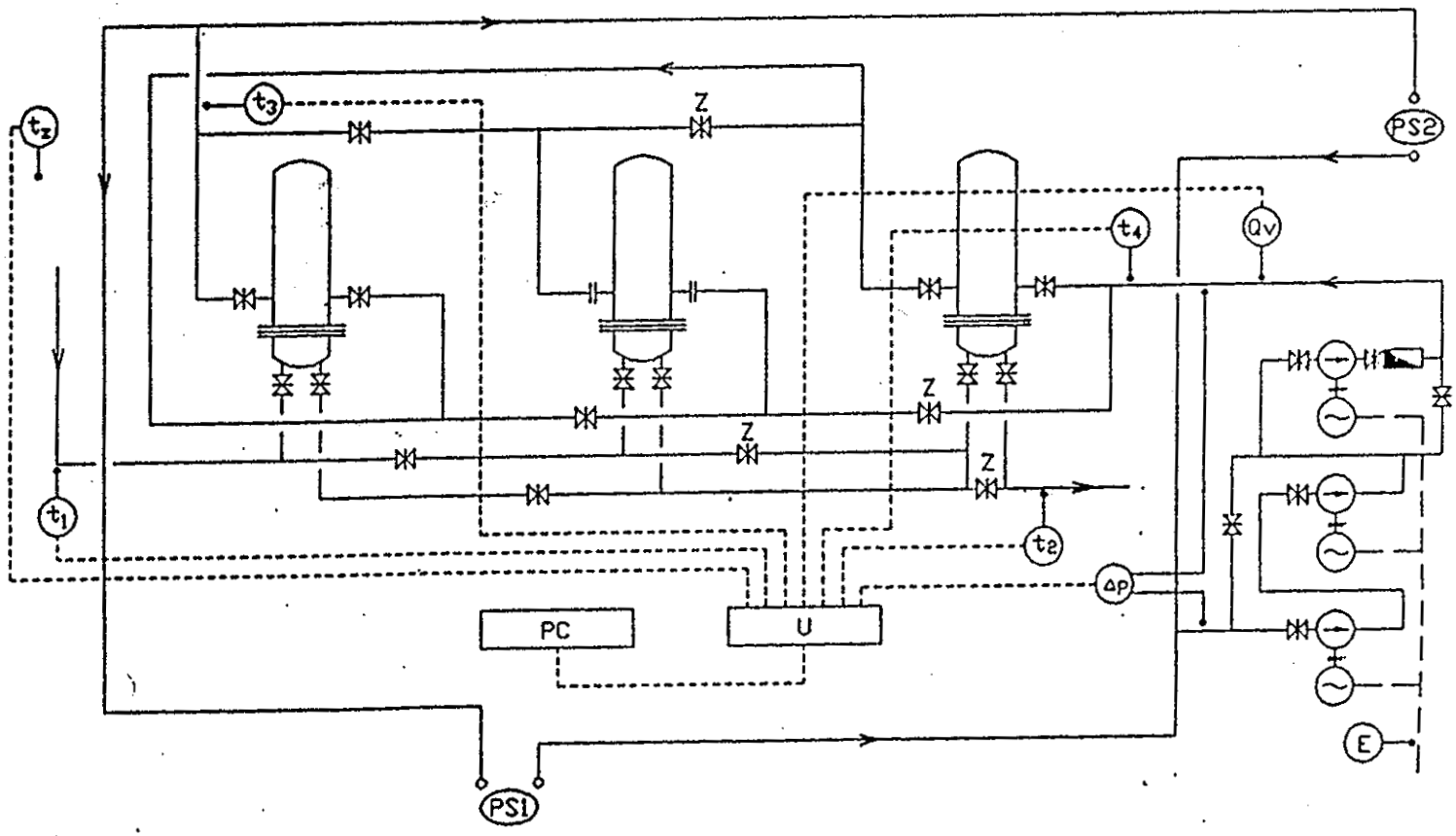
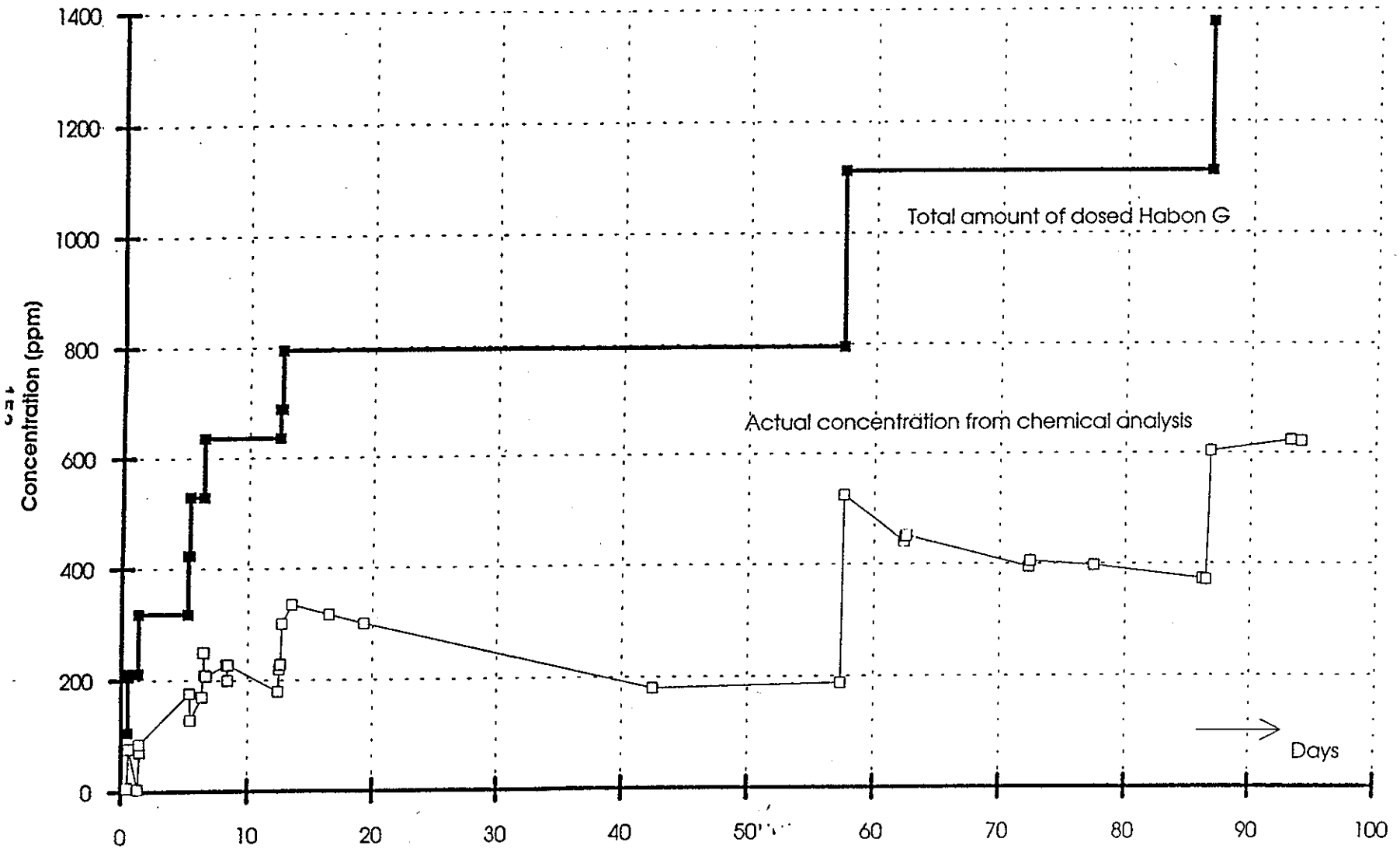
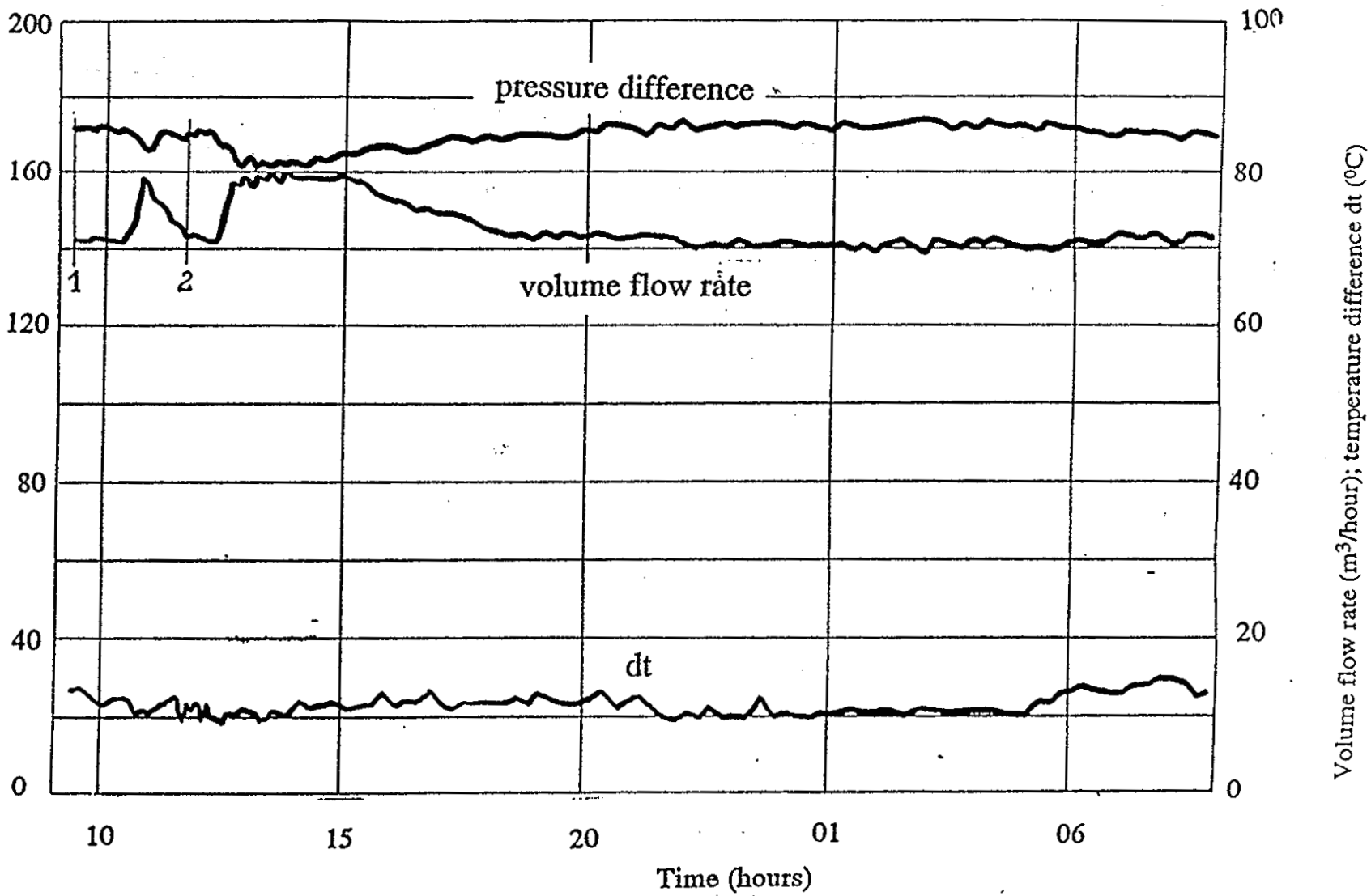


**Fig.3 - Kladno-Krocehlavy Secondary Heating System**




Total volume of 40 m3. Influence of adsorption of heating network on actual concentration of Habon G.

Fig. 4 - Record of Initial Experiments - 1st and 2nd December 1994
Dosing time of Habon G - 10 min




| No. of dosing | Theoretical concentration | Actual concentration |
|---|---|---|
| 1 | 106 ppm | 7 ppm |
| 2 | 212 ppm | 76 ppm |

**Fig. 5: Effect of drag reduction on the secondary heating system characteristics and the performance of the pumps**

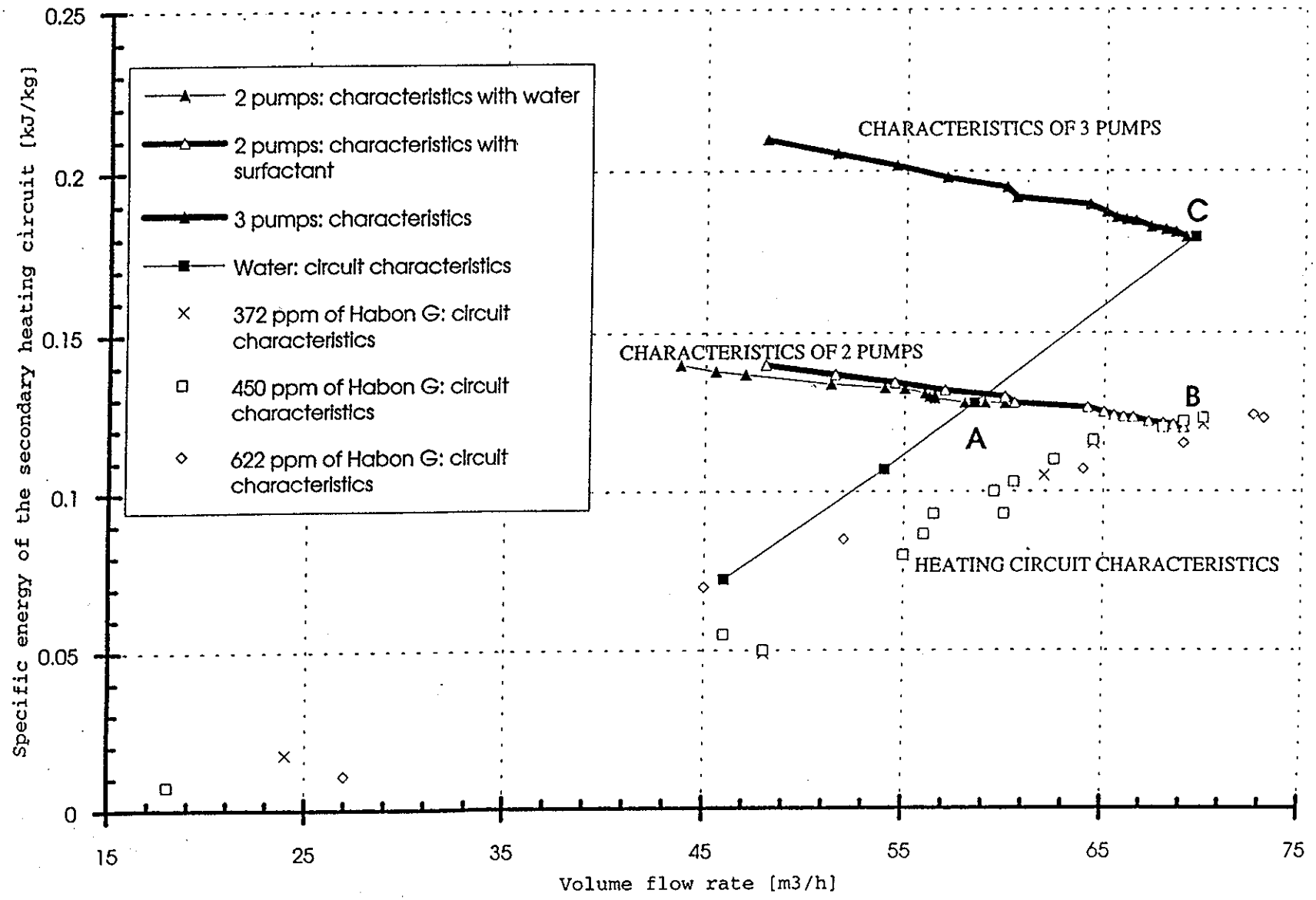





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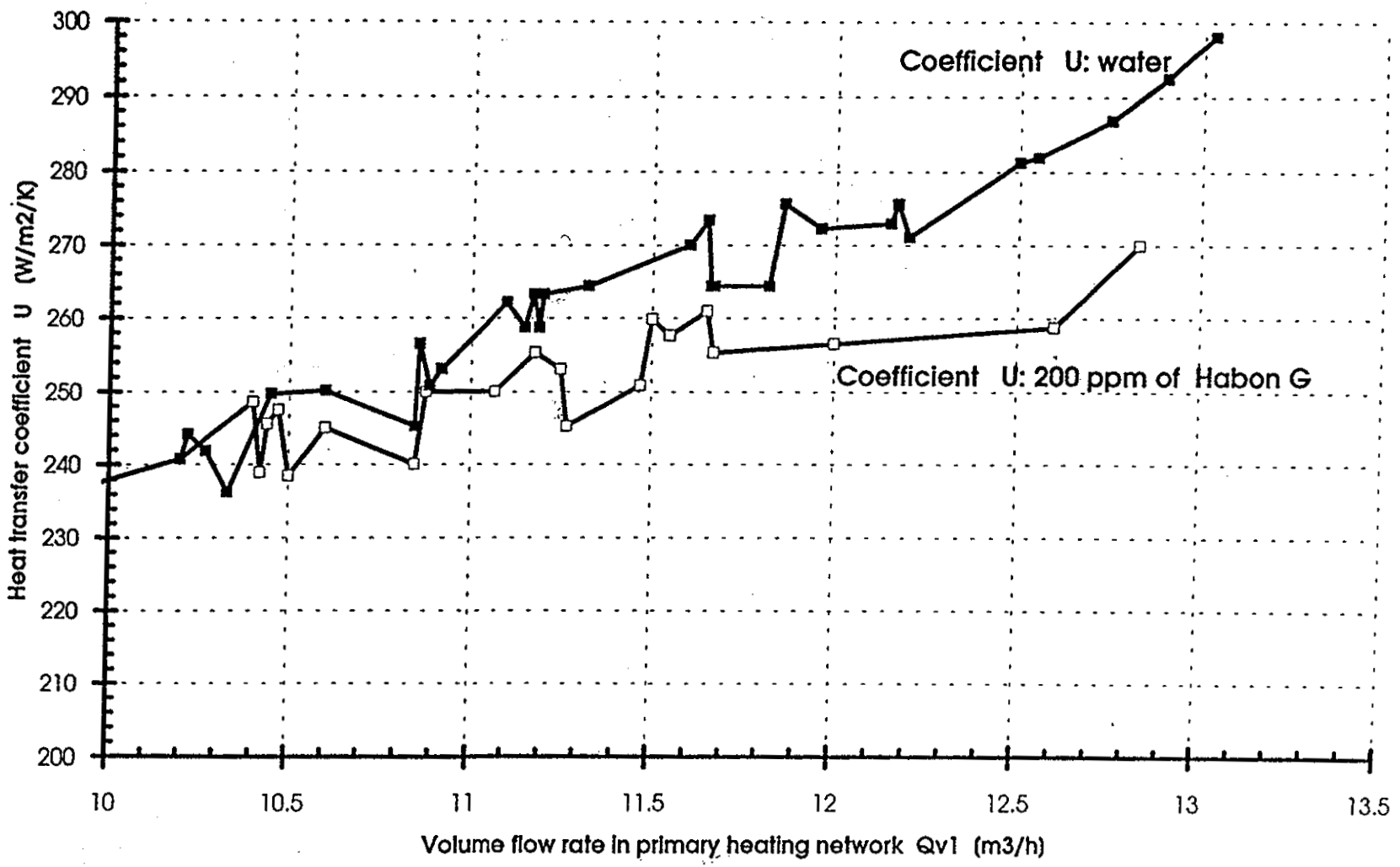
Fig.6: Heat transfer paramaters vs. volume flow rate in primary heating network



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With the compliments of Štern

# Properties of a drag reducing micelle system

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*Abstract*: Rheological properties of a drag-reducing surfactant were investigated with the intention to find out the influence of shear rate and the duration of shear straining on the Shear Induced Structure. The ability of the surfactant to restore the broken drag-reducing network after the decrease of shear stress is well known. This property of reversible change caused by high shear at different flow conditions was compared to a more intensive mechanical straining by means of ultrasound. Observations using electronmicroscope and spectrometer are also presented.

*Key words*: Surfactant – micelle structure – drag reduction – rheological properties

## Introduction

The phenomenon of drag reduction in a turbulent flow in pipes has been studied very thoroughly by a multitude of scientists since it was described for the first time almost 50 years ago. Mainly two types of drag reducing additives were investigated. Polymers were first investigated as effective additives, and later attention was paid also to micelle systems [1–3]. Each additive has specific advantages.

The object of our interest are micelles of some surfactants and their secondary networks which are built up during the flow of such a system in a pipe with respect to the ability of the surfactant to reduce the friction losses in turbulent flow. Cationic surfactants are effective as drag reducers within limited concentration and temperature ranges [4]. This is very important for the possible usage in heating systems [5–6]. It is always emphasized that the surfactants demonstrate "a reversible shear degradation", that is, they suffer a mechanical change of the structure in regions of high shear and regain their drag-reducing effectiveness when the level of shear stress decreases. This behavior is usually ascribed to the breakdown and reformation of the network structure built up from rod-like micelles [7–12]. It is certainly an advantage over the polymeric solutions. The long chain molecules of polymers are subjected to breaking by shear and the degraded polymer has much less ability to decrease the friction than the undegraded one. Moreover, the polymer mechanical degradation is permanent.

## Experimental

The following instruments were used for the measurement and observation of the surfactant solutions: rotational rheometers Rotovisco RV 20 and CV 20 N with coaxial cylinder sensors (Haake, FRG), transmission electronmicroscope Geol JEM 100 B (Geol Ltd., Japan), ultrasonic disintegrator and photon correlation spectrometer.

The first component of the used additive was the commercial product Ethoquad T/13–50 (ET) which is produced by Akzo Chemie America. Its composition is as follows: 50% active tenside-tallow tris hydroxyethyl ammonium acetate, 36% isopropanol, and 14% water. Its molecular weight $Mw$ is approximately 454, specific gravity 0.952 g/cm$^3$ at 20 °C. The second component of the surfactant is natrium salicylate (NA) with $Mw = 160.11$.

Most of our rheological measurements were carried out with the mixture of ratio ET:NA



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= 5 mMol/l of active tenside ET : 7.5 mMol/l of NA (which was 4.54 g/l : 1.20 g/l in total weight).

## Results

### *The evidence of shear-induced structure*

There exist several theories explaining the micelle function in drag reduction in the turbulent flow. After Hoffmann et al. [9], the rod-like micelles make up a lattice in which the rods are statistically distributed. Their orientation is arbitrary, but when sheared they become ordered parallel with the stream. Such an oriented structure grows from small clusters to the entire flow field with the increasing rate of shear. This network dampens the turbulence. It is broken up when a certain rate of shear is exceeded and the drag-reducing effect is lost. When the rate of shear is diminished again, the oriented lattice reshapes.

Another theory [11, 12] states that the supermolecular structure is built up from single micelles. The rod-like micelles bounce against one another, join and form long parallel chains in the flow field. A similar hypothesis was expressed in [8], with the presumption that the micelles are flexible and that the velocity fluctuations perpendicular to the flow are dampened by long micelle chains. With high values of rate of shear the chains break, but when the rate of shear is decreased again, they reform.

Orthokinetic aggregation of colloidal particles is well known. For example, the growth of latex particles caused by shearing was studied by Husband and Adams [13]. This effect occurred at rather high shear rates above $10^3\ s^{-1}$. A similar mechanism may take place in aggregation of micelles up to a critical shear rate when the aggregates break. The computer simulation for aggregation of sphere-shaped colloids at low shear rates was made in [14] with the result of steep increase of viscosity (when plotted against shear strain at constant shear rate) at the beginning and achievement of the constant viscosity for higher values of shear strain. Our measurement (not presented here) of time dependence of viscosity at constant low shear rates proved the quick viscosity change at the beginning and no time dependence after a certain time.

Shear-induced phase transitions were described by several authors [4, 10–12]. The shear-induced structure (SIS) as a result of growing shear strain can also be very well observed by means of rheological measurements. The appearance of SIS in water solutions of Etna with different concentrations is presented in Fig. 1. As can be seen, the viscosity of the solutions decreases at first with the rising rate of shear (pseudoplastic behavior), and at a certain critical value of the rate of shear a sudden increase of viscosity is encountered which is caused by the formation of SIS. We conclude also that the critical value of the rate of shear for the formation of the SIS is smaller with the smaller tenside concentration.

The effect of temperature on SIS is shown in Fig. 2; the critical value of the rate of shear decreases with increasing temperature as well as with decreasing concentration (Fig. 1). In this respect the surfactant Etna differs from some other described micellar systems [4, 15]. Increasing the rate of shear beyond the region of the SIS origin (above about $1100\ s^{-1}$) there appears again a decrease of viscosity. This region can be called the second region of pseudoplastic behavior. Repeated measurements unambiguously showed that the structural changes due to temperature straining are reversible. When measuring the temperature

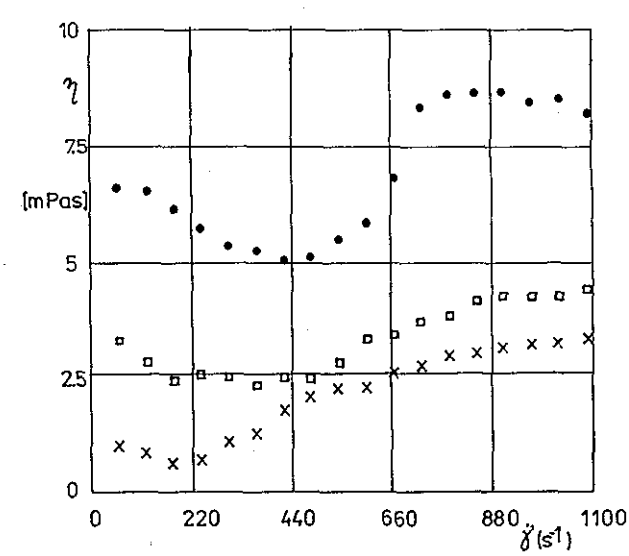


Fig. 1. Viscosity curves of Etna solutions with the total weight ratio ET : NA = 3.78 : 1 and total weight concentrations: ● 5.74 g/l; □ 4.305 g/l; × 2.153 g/l

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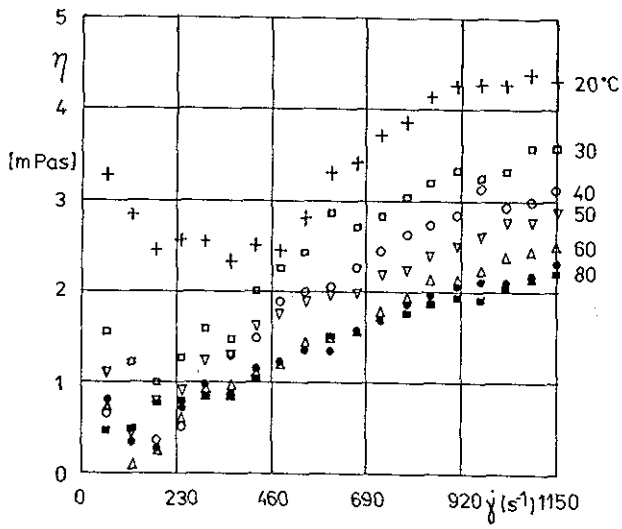


Fig. 2. The temperature dependence of viscosity of Etna solution with total weight concentration 4.305 g/l

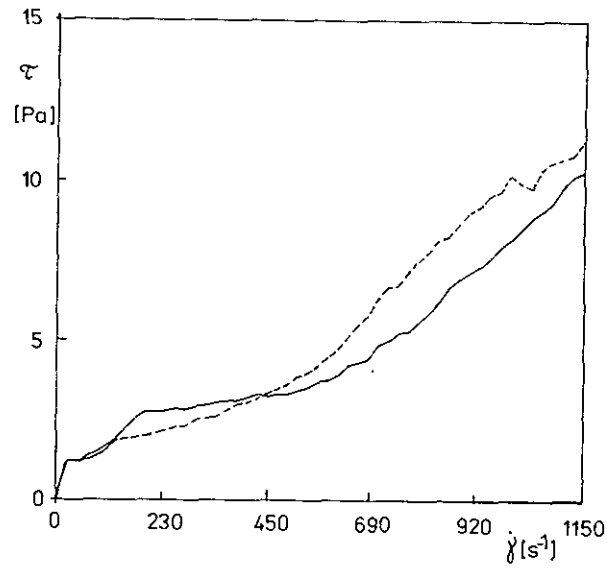


Fig. 3. The flow curves of Etna solution (5.74 g/l) at 20 °C. Solid line: fresh sample; dashed line: after 60 min shearing at $\dot{\gamma} = 200\ s^{-1}$

dependence of the flow curves, control measurements at 20 °C were performed after each temperature step. No changes in the flow curves at 20 °C were observed, thus indicating the good stability of the surfactant with respect to temperature history (within the range 20–80 °C).

Not only the magnitude of the rate of shear influences the rheological properties of the surfactant, but also the duration of application of the shear rate reveals similar effect. To prove this two flow curves measured with Rotovisco RV 20 are drawn in Fig. 3, the one of fresh sample and the second flow curve measured after straining the sample for 60 min by constant rate of shear $\dot{\gamma} = 200\ s^{-1}$. As can be seen, the flow curve of the strained surfactant reaches higher shear stresses than those of unstrained surfactant. However, repeated measurements of flow curves showed that the structural changes due either to small shear straining or its short duration are reversible as no permanent changes in the flow curves were observed.

*Breaking of the secondary micellar structure*

As we mentioned above, there is a generally accepted idea that the secondary micelle network system breaks at a certain level of shear strain but that it reshapes again when the strain loaded on the solution decreases. The system is able to regain the drag-reducing properties. However, this concept does not apply to any mechanical strain which we proved by measurement of the influence of ultrasound application [16] on the rheological properties of water solution of Etna.

The solution (of 0.574% weight concentration) was strained by ultrasound in a 100 ml beaker for 2 or 8 min. The steady shear viscosity curves were measured before and directly after the exposure to ultrasound and again after 3 days of rest. All measurements were done at the same temperature 20 °C and the results are shown in Fig. 4. The influence of ultrasound is easily seen there; the viscosities of solutions after the straining are lower than those before straining. Figure 4 proves that the secondary micellar structure changes due to ultrasound load. This change is not reversible because the flow curve measured 3 days after straining is almost the same as the one measured immediately after straining. The flow curves of the strained sample at different temperatures are shown in Fig. 5. In comparison with the temperature influence on the flow curves of the unstrained surfactant, the strained sample viscosities are always lower.

The acoustic power at the top of the concentrator of the ultrasound generator was 40 to 50 W and the energy input to the solution during 2 min

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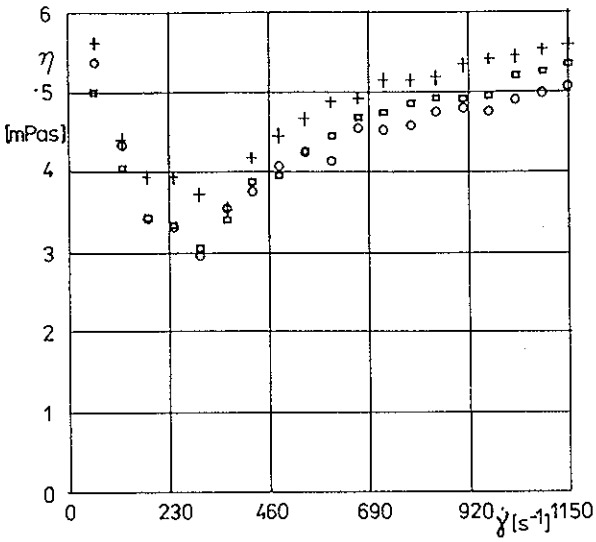



Fig. 4. The viscosity curves of Etna solution (5.74 g/l) at 20 °C. + fresh sample; □ after 2 min straining by ultrasound; ○ after 8 min straining by ultrasound

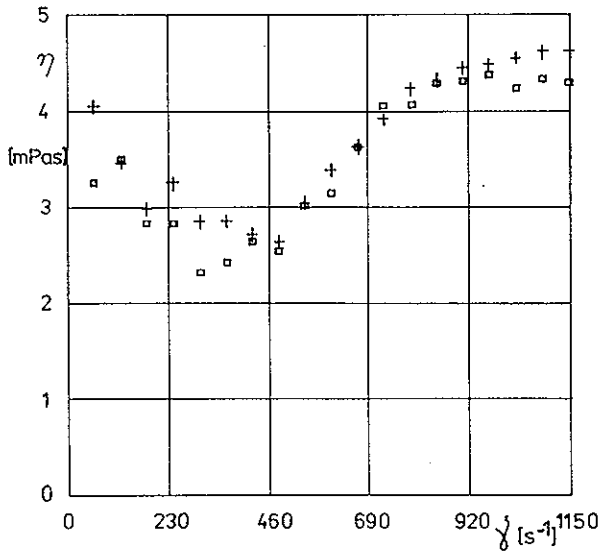



Fig. 6. The viscosity curves of Etna solution (4.305 g/l) at 20 °C. – fresh sample; □ after 2 min straining by ultrasound

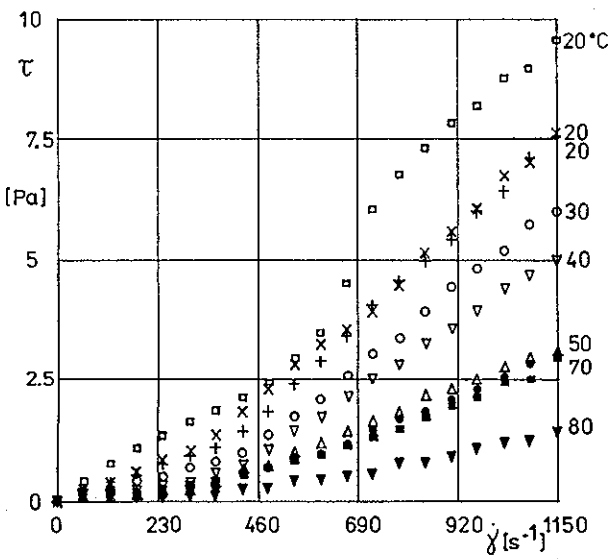



Fig. 5. The flow curves of Etna solution (5.74 g/l). □ fresh sample; + sample measured immediately after straining by ultrasound; all other symbols are temperature-dependent flow curves of the strained sample measured after 3 days' rest

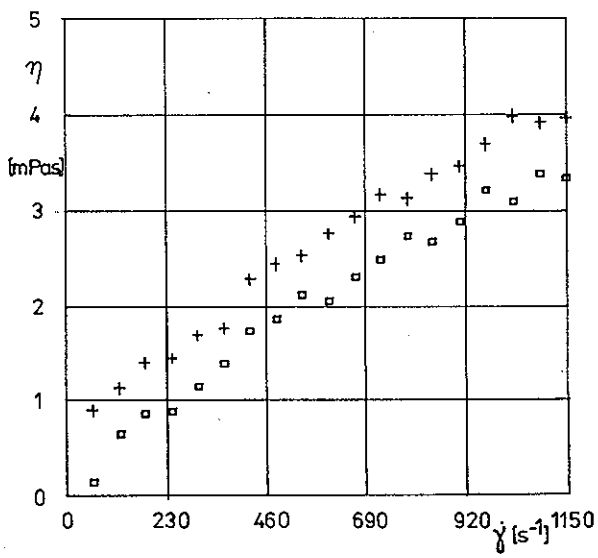



Fig. 7. The viscosity curves of Etna solution (2.87 g/l) at 20 °C. – fresh sample; □ after 2 min straining by ultrasound

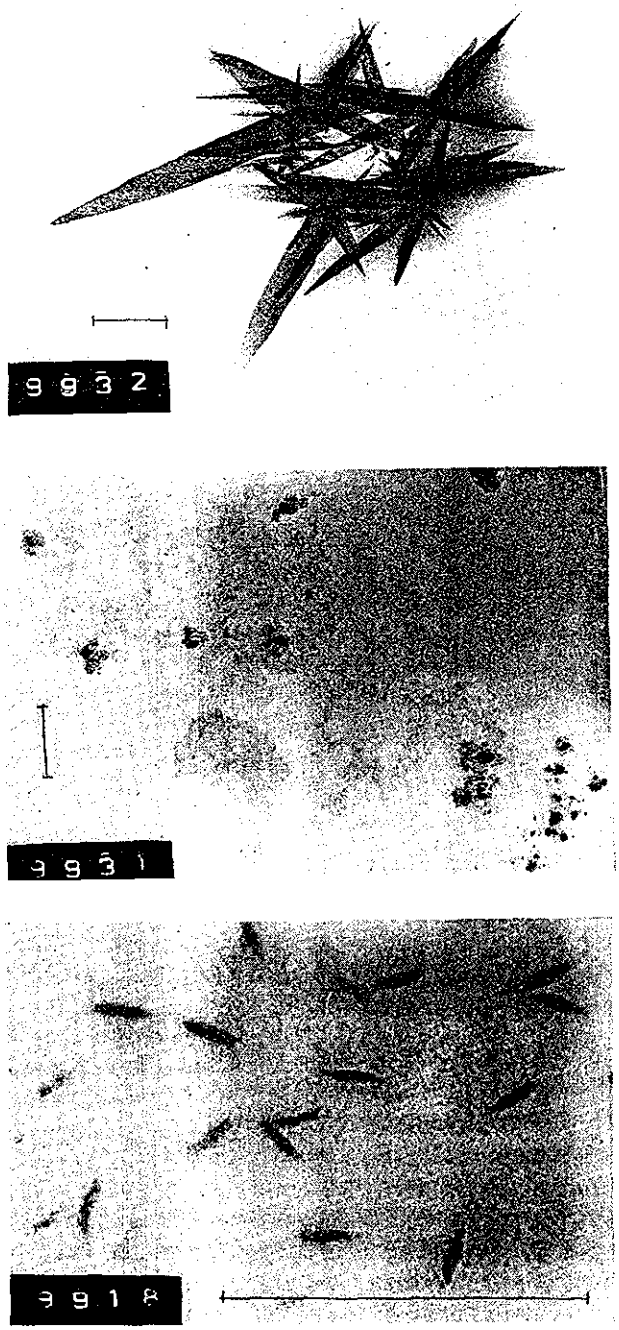
was 4800 Ws, which equals the specific work of 32 Ws/g. Part of energy input is the heat loss.

The viscosity curves are shown also in Figs. 6 and 7. The viscosity after ultrasound application is always smaller than the one before application. Longer application decreases the viscosity even more (see Fig. 4). The flow curves after 3 days' rest do not change. This proves that the partial permanent change of the micelle network by means of very intensive mechanical straining is possible.

The influence of other types of mechanical straining is now being investigated.

*Observation in electronmicroscope*

One author has observed [17] the shape of particles of a surfactant Sean, which is a mixture of carbo ethoxy pentadecyl trimethyl ammonium bromide with 1-naphthalene. This surfactant proved to be a very effective drag reducer at room temperature [8]. A similar procedure as in [17] was used in the observation of the surfactant Etna presented here.



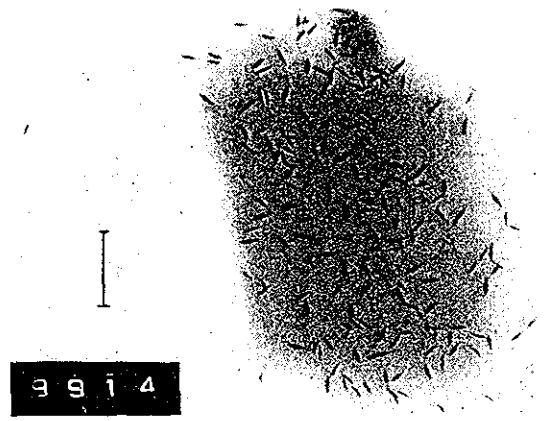




Fig. 8. Electron micrographs of observed dried microparticles of Etna solutions. The line segment equals 1 $\mu$m. a) and b) concentration 0.544%; c) and d) concentration 0.1435%.

Observation was done in a transmission electronmicroscope Geol JEM 100 B. The applied voltage was 60 kV. The microscopic preparations for observation were made in such a way that the solution was either dropped or finely sprayed on the underlying collodion film. In Fig. 8 are shown different shapes of observed particles from the dried surfactant Etna. The dried samples do not necessarily correspond to the true wet micellar or network structure. Both shape and size of oberved objects are remarkably similar to the ones from drag-reducing surfactant Sean [17].

*Hydrodynamic radius of micelles*

The size of micelles at rest was measured by means of quasielastic light scattering. The distributive functions of hydrodynamic radius $R_H$ of micelles were obtained by Laplace transform from autocorrelation functions of intensity of the scattered light. Three different concentrations of Etna were tested as shown in Fig. 9. Only a very small influence of concentration on average values of $R_H$ was determined.

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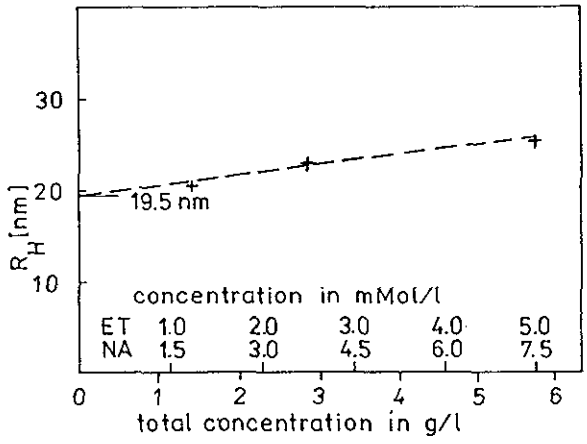


Fig. 9. The hydraulic radius $R_H$ of micelles measured by means of light scattering and its dependence on concentration of Etna solutions.

## Conclusion

Cationic surfactants are effective as drag reducers. It had been found that the micelles form the SIS at a certain value of rate of shear. This structure was confirmed by the rheological investigation of the water solution of Ethoquad T 13/50 with natrium salicylate.

It is known that the secondary network structure, which is responsible for the drag reduction, suffers changes in regions of high shear, and the network reshapes again when the level of shear stress decreases, thus demonstrating the ability of reversible change of network. It was proved that this surfactant may also display a permanent change of viscosity curve by ultrasound application.

We have pointed out a remarkable similarity between the shape and the size of observed dried structures of two different surfactants: carbo ethoxy pentadecyl trimethyl ammonium bromide with 1-naphtalene and tallow tris hydroxyethyl ammonium acetate with natrium salicylate. Both surfactants are effective drag reducers. The size of micelles of Etna at rest as measured by light scattering is approximately 0.05 $\mu$m.

*Acknowledgement*

Financial support of this work by grants from the Agency for International Development, Washington, D.C., and Czechoslovak Academy of Sciences, Prague, is gratefully acknowledged. We thank Dr. Starý from the Microbiological Institute, Dr. Malinský from the Czech Technical University and Dr. Štěpánek from the Institute of Macromolecular Chemistry for collaboration in the research.

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# FLOWS OF SURFACTANT DRAG REDUCERS FOR DECREASING OF PUMPING ENERGY DEMAND IN DISTRICT HEATING SYSTEMS

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Czech Technical University, Prague, Czech republic
Jacques L.Zakin
Ohio State University, Columbus, U.S.A.

## 1. INTRODUCTION

The world-wide use of district heating for space heating is increasing as it can provide an environmentally acceptable low-priced supply of energy. By utilizing waste heat from cogeneration plants, it increases the efficiency of fuel utilization under controlled and monitored operating conditions.

The high investment costs of the heat transport and distribution network with only partial utilization during the year make district heating systems less attractive so that it is important to reduce their investment and operating costs. A promising possibility is to use surfactant drag reducing additives in district heating systems. These additives can reduce power requirements in an existing system or reduce investment costs by utilizing smaller diameter distribution systems and/or smaller capacity pumps.

Many cationic surfactant-counterion systems form microstructures which are highly effective in reducing turbulent friction up to a critical shear stress above which the structures are degraded and fluid behaves as a Newtonian fluid with no friction reduction. If the stress is reduced below the initial point the structure is reformed and the friction reducing ability is restored [1].

## 2. EXPERIMENTAL

Measurements were made on two quarternary ammonium-counterion systems. Habon G (cetyl dimethyl hydroxyethyl 3-hydroxy 2 naphtoate) was donated by Hoechst GmbH, F.R.G. Ethoquad 0/12 (tetradecyl trimethyl ammonium bromide - production of AKZO Chemicals, U.S.A.) was used at a concentration of 5.0 millimoles per liter with an excess of sodium salicylate (12.5 millimolar).





The test sections for the drag reduction measurements on Habon G in the Prague laboratory consisted of straight stainless steel tubes, of differen diameters (4, 6, 10 and 20 mm) each 5 m long with its own flowmeter and pressure measuring system. The construction of the laboratory pipe test loop system permits the drag reduction measurements to be carried out in two ways For ordinary evaluation of drag reduction once through measurements were made. The second possibility was to connect different sizes of tubes into smal simple networks (parallel tubes) to evaluate the influence of the network on drag reduction. Control tests were run on Prague tap water (used for all tests) and compared with the Prandtl-von Kármán friction factor relation. Results for water at all temperatures showed excellent agreement with values predicted from this equation. All surfactant additive experiments were carried out at three temperatures - 20, 40 and 60° C. The duration of each test was 15 hours.

The tests on the Ethoquad 0/12 were carried out in the Ohio State University test loop in stainless steel tube of D = 6.2 mm and test section length of 0.5 m.

## 3. RESULTS AND DISCUSSIONS

The data for all experiments showed strong temperature influence, i.e. as the temperature increased the maximum solvent Reynolds number at which significant drag reduction occurred also increased up to a maximum critical temperature. For Ethoquad 0/12, Fig.1 shows that drag reduction is observed up to higher solvent Reynolds numbers as temperature is increased until the maximum effective temperature of 40° C for this system is reached.

One of the most interesting effects observed in studies of drag reduction by polymers is the "diameter effect". This refers to the fact that for a given fluid and Reynolds number, the friction (except in the maximum drag reduction asymptote regime) is a function of the diameter of the pipe, i.e. for a given polymer and concentration, drag reduction at a given Reynolds number is greater in small pipes. This is, of course, not the case with Newtonian fluids for which the pipe diameter is already taken fully into account by the Reynolds number.

Drag reduction data for 0.05 % Habon G solutions are shown in Fig. 2. for five different pipe diameters. At high values of the wall shear stress, the solution becomes ineffective for drag reduction and behaves like the solvent. Because of the high wall shear stresses in the small tubes, the influence of pipe diameter is strong, i.e. with increased pipe size, drag reduction increases as does the range of Reynolds number in which the surfactant is effective because the critical wall shear stress for mechanical degradation is exceeded in the small tubes at moderate Reynolds numbers.. This is contrary to results for polymer additives where drag reduction increased with decrease in pipe size.

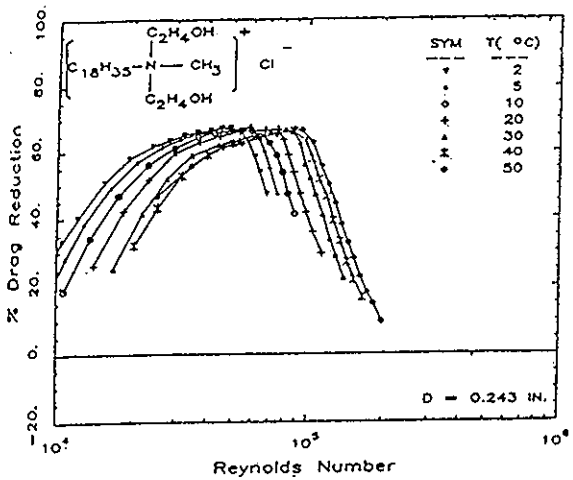


Fig.1 - Influence of temperature om drag reduction efficiency; surfactant Ethoquad 0/12 + NaSal, concentration 5.0/12.5 mM; D = 6.2 mm

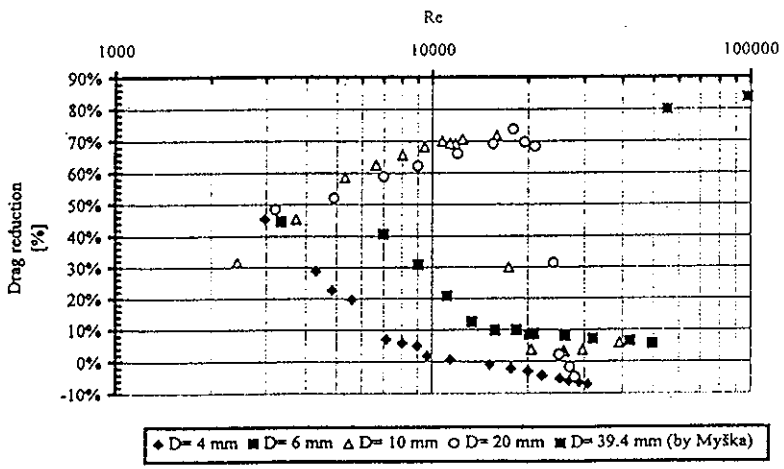


Fig.2. - Changes of drag reduction in different pipe sizes under isothermal conditions, Habon G of 0.05 %, t = 20 °C

For all surfactants at all concentrations that were studied in single tubes, the results fall into two distinct groupings; one of the smaller size tubes: 4 and 6



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mm, and the other, the larger tubes 10 and 20 mm (it is also possible to include the results from the large pipe D = 39.4 mm, in this group). In the smaller tubes the critical wall shear stress was reached at low Reynolds number but sharp changes in drag reducing effectiveness in a narrow range of Reynolds number were not observed. But in the cases of the 10 and 20 mm pipe sizes it was possible to see typical surfactant drag reduction additive behaviour in which the stress causes breakdown of microstructure of the micelles when the shear stress exceeds a critical value with a relatively sharp increase in pressure.

It is not clear why drag reducing data fall into two groupings. One possible explanation is related to the differences in the shapes of the velocity profiles with surfactant and without surfactant [2]. Velocity profiles in drag reducing micellar solution systems are quite flat in the central region. In the smaller 4 and 6 mm tubes, the microstructure size may be large relative to the distance from the wall at which the expected steep portion of the profile should lie, thus interfering with the development of the profile.

Very interesting results appeared when drag reduction effect of Habon G in a simple network was studied, Fig.3. The results show that the pipe network has a strong influence on drag reduction in the 4 mm tube, i.e. drag reduction zone and the effect are increased in the 4 mm tube when it is in parallel with the 10 mm tube, in comparison with a single 4 mm tube system. The drag reduction effect in the 10 mm tube is constant, however, without change compared with the values in a single tube. The diameter ratio between the tubes connected in the system should be greater than 2.5 for a change in drag reduction effectiveness of the small tube. For smaller ratios between the tubes, improvement in the drag reduction in the smaller tube was not observed.

To better understand the influence of a complicated network system on overall drag reduction, a field test was run on the secondary heating network of one typical Czech housing estate: Kladno-Kročehlavy [3]. During the field experiment the concentration of Habon G was incrementally increased 10 times. Results from analytical investigations of concentration showed a large difference between the actual concentration and the calculated concentration based on the amount of injected additive, which was caused by adsorption of surfactant on the walls. The initial difference was large with only 7 ppm in the system despite dosing at a calculated level of 106 ppm. After the last dosing, the calculated concentration was 1378 ppm and the actual concentration was 623 ppm. Only a minor part of this difference can be attributed to leakage.

Normal operation of the secondary system in Kladno-Kročehlavy used three centrifugal pumps which provided an adequate supply of hot water to the

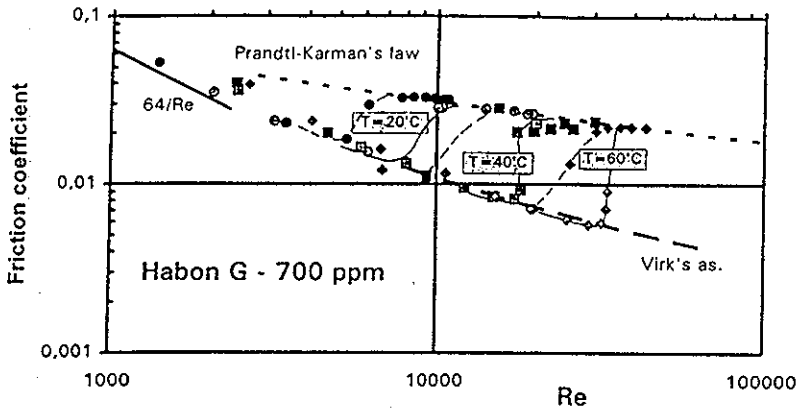


Fig.3.- Comparison of DR effect in 4 mm pipe :
a) — — — single tube system,
b) ——— simple network ( 4 + 10 mm )

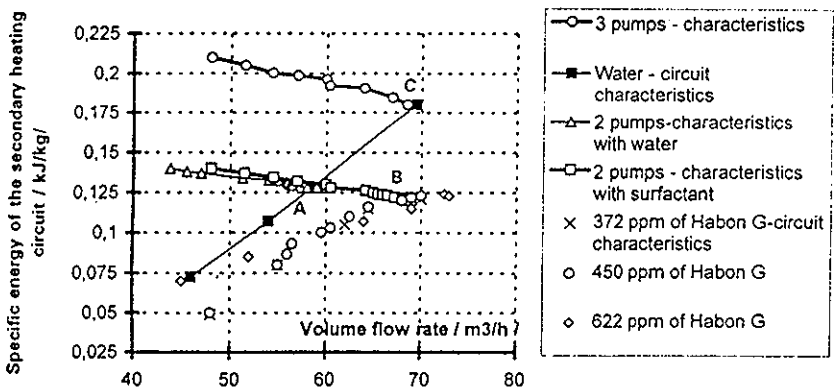


Fig.4. - Drag reduction effectiveness on the secondary heating systems characteristics and the performance of the pump

radiators. When concentration reached ca. 400 ppm, total reduction of specific pumping energy reached 30 % which was equivalent to the power provided by one of the pumps. At that point the third pump was switched off and the whole secondary system operated with only two pumps without any difficulties in spite of very cold weather during two weeks of the heating season in the winter 1994. Fig.4 shows details of the results including changes in the characteristics



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of centrifugal pumps and the specific energy demand of the secondary heating system with different Habon G concentrations. The maximum decrease in specific energy, 40.8 %, was achieved at a concentration of 450 ppm of Habon G. When actual concentration was increased above 372 ppm, no significant increase in drag reduction was observed. Thus, the optimum actual concentration for such a system is 400 - 450 ppm. At this concentration of Habon G, total drag reduction was very stable for several weeks.

## 4. CONCLUSIONS

1. The introduction of cationic surfactants to pipe flow systems causes a significant reduction of pressure loss in turbulent flow.
2. For the surfactants, the influence of tube diameter on drag reduction is strongly affected by wall shear stress which is higher in small diameter tubes. As a result, the level of drag reduction at high Reynolds numbers is larger for large tubes which have not exceeded the critical wall shear stress. In polymer solutions, drag reduction is generally greater in small tubes.
3. In networks flows through small and large tubes in parallel in which the diameter ration exceeds 2.5 : 1, drag reduction effectiveness in the small tube is surprisingly extended to higher Reynolds numbers compared with flows through only small tube.
4. Drag reduction is observed up to higher solvent Reynolds numbers as temperature is increased until the maximum effective temperature is reached.
5. During experiments in a closed loop pipeline system no mechanical or ageing degradation effects were observed as are found with polymer additives.

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# Viscoelasticity of a Surfactant and Its Drag-Reducing Ability

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**Abstract.** There is considerable interest in the use of viscoelastic cationic surfactant-counterion mixtures in district heating and cooling systems to reduce pressure losses. A recent field test in a secondary system near Prague showed a 30+% reduction in pumping energy requirements.

We have studied a number of commercial surfactants and we report here results of rheological, drag reduction and turbulence measurements on Arquad 18-50 (octadecyl trimethyl ammonium chloride (AR 18)) with an excess of sodium salicylate (NA). The concentration studied was 1.6 mM AR 18 and 4.0 mM NA which is about one third the concentration for excellent drag reduction in this surfactant's effective temperature range 30–90°C.

Viscosity, $\eta$, vs. shear rate, $D$, first normal stress difference, $N_1$, vs. shear rate, drag reduction (as pressure drop, $i = \Delta P/1$) vs. average velocity, $U_{ave}$, in a 39.4 mm tube for AR 18, and turbulence intensity data for three drag reducing surfactants are reported.

Of particular interest are the generally low turbulence intensities in all three directions which correspond to reduced heat, mass and momentum transfer rates compared to water, and the existence of large normal stress differences at 20°C for AR 18, a temperature at which no drag reduction occurs with this surfactant, indicating that normal stress effects do not correlate directly with drag reduction.

The effect of time of pumping on increasing drag reduction demonstrates that this factor overwhelms the expected increase in drag reduction as temperature is raised from 18–19°C to 40.5°C.

**Key words:** cationic surfactant drag reduction, shear induced structure, viscoelasticity, turbulence intensities

## Introduction

In the past several years, there has been considerable interest in the use of low concentration, viscoelastic cationic surfactant-counterion mixtures for reduction of pumping energy losses in district heating and district cooling systems. Field tests in Germany [1], in Denmark [2], in Ukraine [3], and in the Czech Republic [4] were made. For example, in a small secondary system in Kladno-Kročehlavy near Prague, pumping energy requirements were reduced by one third when a surfactant drag reducing additive was injected into the loop [4]. Even larger reductions have been observed in primary systems having long straight stretches of pipe [1].

There are, however, a number of uncertainties regarding how surfactant drag reducing additives affect drag reduction and turbulence structure, and what influence shear induced structure (SIS) and other rheological properties such as normal stresses have on drag reduction.

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In the experimental results reported here, measurements of shear viscosity, first normal stress differences, turbulent pressure drop and turbulent intensity were made on a commercial surfactant Arquad 18-50 (AR 18 below) with an excess of sodium salicylate (NA below). This mixture shows very strong normal stress behavior to at least 70°C. The results are compared with water and other drag reducing surfactants Ethoquad T13/50 (ET 13 below) and Habon G.

The experiments were carried out at convenient temperatures to determine trends in drag reduction, rheological behavior and turbulence intensities. Once these are understood, they can be extended to higher or lower temperatures characteristic of district heating or cooling systems. A study of the heat transfer characteristics of drag reducing surfactants is also underway.

## Experimental

Arquad 18-50 ($C_{18}H_{37}$-N-$(CH_3)_3Cl$) contains 50% surfactant dissolved in a mixture of water and isopropanol and is easily dispersed in water as is Ethoquad T/13-50 (Tallow-N-$(C_2H_4OH)_3Ac$) which also contains 50% surfactant. Both were donated by Akzo Chemical Co. Compared is also Habon G [$C_{16}H_{33}$-N-$(CH_3)_2$-$(C_2H_4OH)$·(3-OH-2 naphtoate)] which was donated by Hoechst AG and contains 53% active matter. All concentrations of surfactants given below are concentrations of active matter in distilled water unless noted otherwise (which is a well defined solution for rheological measurements).

Measurements of shear viscosity and first normal stress differences were made with a Haake rheometer Rotovisco RV 20 and CV 20 N with a 2° cone-plate system and also with a coaxial cylinder system [5].

Pressure drop measurements were made in a 39.4 mm ID glass tube recirculation system with a straight test section 5.8 m long. The facility was described in [6]. The solution was kept at a constant level in an overflow tank. The test section itself was approximately 2 m long and was provided with open manometers. A rectangular box made of thin glass enveloped the tube in the middle of the test section. The box was filled with water in order to suppress undesired reflections of the laser beams. The rate of flow was controlled by a level gauge connected with electric timer installed in a collecting tank. LDA optics assembled from standard Dantec components were mounted together with the Argon-ion laser (Coherent INNOVA 70) on a remote controlled $xyz$ support. Evaluation of the forward scattered Doppler signals was performed by Dantec Burst Spectrum Analyzer BSA which makes possible more precise measurements than Particle Dynamic Analyzer which we used in our earlier measurements. The total length of tubing was approximately 16 m, the total system volume was 520 liters. Circulation was by a centrifugal pump.

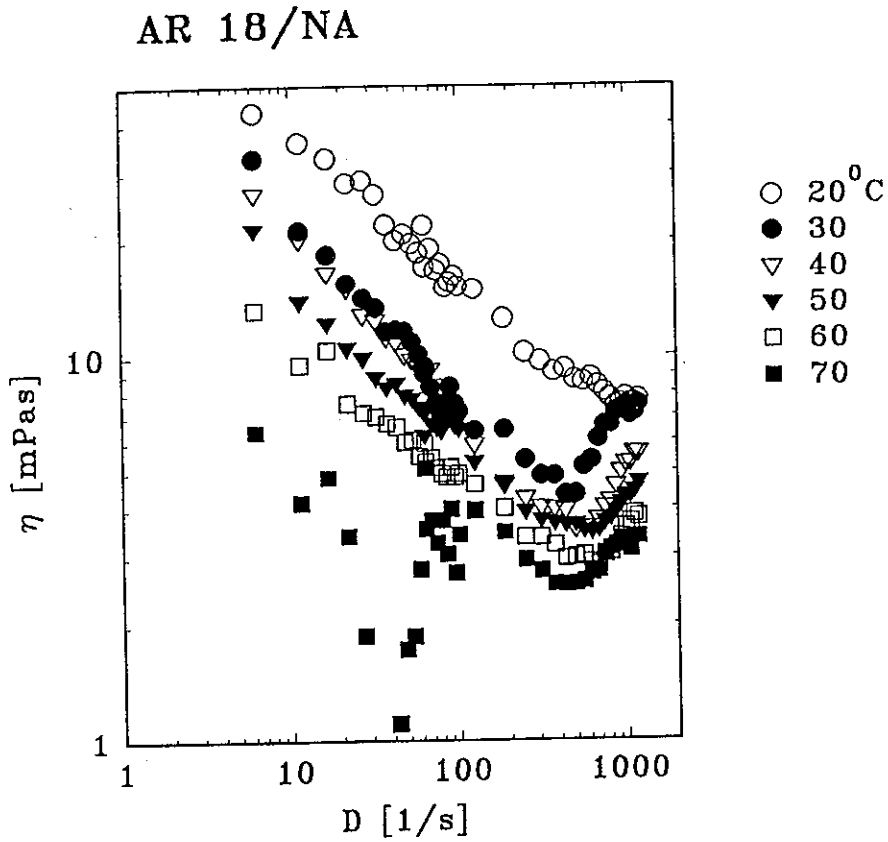


*Figure 1.* Temperature dependence of apparent viscosity of AR 18/NA solution on shear rate in the range of 0–100 $s^{-1}$. The concentration in distilled water is 5/12.5 mM, coaxial cylinder system.

## Results and Discussion

### APPARENT VISCOSITY

Figure 1 shows the effect of shear rate on the apparent viscosity of the 5/12.5 mM AR 18/NA mixture in the temperature range 20–70°C. While viscosities are high at shear rates below 10 $s^{-1}$ (when there is the highest degree of micelle entanglement), it decreases with shear rate until about 400 $s^{-1}$, except for 20° for which the viscosity reaches equilibrium. At higher shear rates, the apparent viscosity increases for all temperatures except 20°C. The decrease of viscosity must be due to the gradual organization of the flow structure and alignment of micelle chains with the direction of the flow. This behavior was confirmed also by birefringence and extinction angle measurements in surfactants, see e.g. [7–9]. In some surfactants, however, the equilibrium viscosity holds up in the whole range of shear rates since it has been achieved (we have found this behavior e.g. in Habon G solutions)

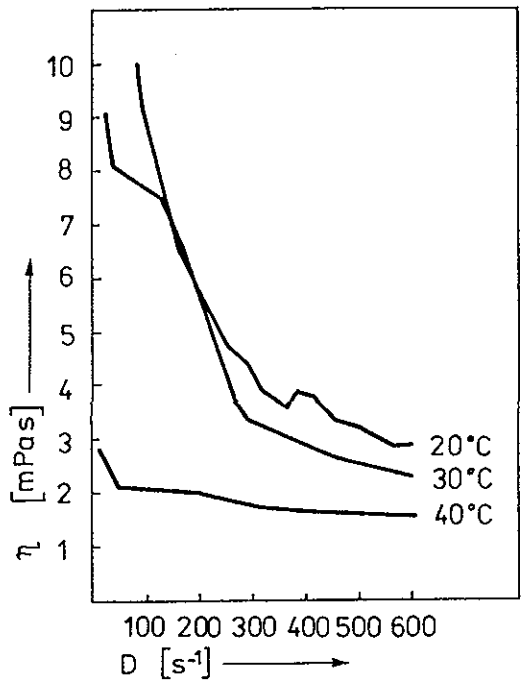


*Figure 2.* Temperature dependence of viscosity of AR 18/NA solution on shear rate in the range of 0–600 $s^{-1}$. The concentration in tap water is 1.6/4.0 mM, the sample was taken at the beginning of drag reduction experiments. Measured in cone-plate system.

but here in Figure 1 we see that the apparent viscosity of this solution increases slightly with shear rate except for 20°C. This shear thickening effect is a result of build-up of a shear induced structure (SIS). This phenomenon has been described previously, for example in [7, 11, 12]. Another manifestation of this effect will be seen in drag reducing data as shown in Figures 6 and 7.

The AR 18/NA solution diluted to 1.6/4 mM in tap water yields only decrease in viscosity in the shear rate range 0–600 $s^{-1}$ (Figure 2) showing a tendency to an equilibrium viscosity which would suggest an achievement of a stable structure at a certain shear rate. The viscosity of this solution which was used in the turbulence measurements and which exhibited also great drag reduction, increased 5–10% by the recirculation during experiments. Increase in viscosity with time of pumping or preshearing was also observed with other surfactants [10]. This again suggests that a slight change in the micelle network occurs even during shearing by centrifugal pumps.

## FIRST NORMAL STRESS DIFFERENCE

AR 18 with excess NA in distilled water is a very effective drag reducing additive at concentrations of 5 and 12.5 mM, respectively, from 30 to 90°C [13]. Based on

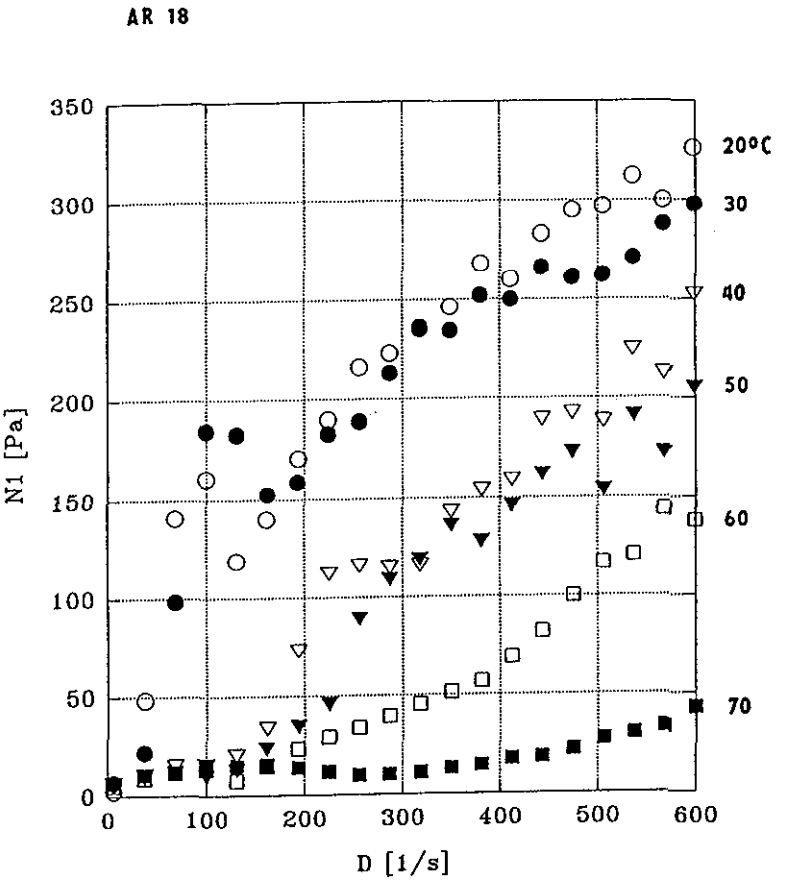


*Figure 3.* Temperature dependence of first normal stress difference $N_1$ on shear rate $D$. AR 18/NA solution with 5/12.5 mM concentration in distilled water. Symbols as in Figure 1.

observations of recoil and on first normal stress difference shown in Figure 3, it is highly viscoelastic up to at least 70°C. Its normal stresses at 30°C and above exceed those of ET 13, Arquad 16-50 ($C_{16}H_{33}$-N-$(CH_3)_3Cl$) and Ethoquad 0/12 (Oleyl-N-$(CH_3)(C_2H_4OH)_2Cl$) – all of which are products of Akzo Chemical – at the same 5 mM surfactant and 12.5 mM NA concentrations. Accordingly, to check the effects at lower concentration, experiments were carried out at AR 18/NA = 1.6/4.0 mM. This lower concentration was chosen in order to see how the surfactant, though still highly viscoelastic, behaves when it does not already possess the strength to maximum drag reduction efficiency as at abundant concentration. The influence of both distilled and tap water is checked.

First normal stress differences, $N_1$, vs. shear rate, $D$, at this lower concentration are shown in Figure 4 for 20 to 40°C. The sample was taken at the beginning of experimenting in the recirculation system where tap water was used. The values of $N_1$ are greater at the lower temperature and rise monotonically with shear rate

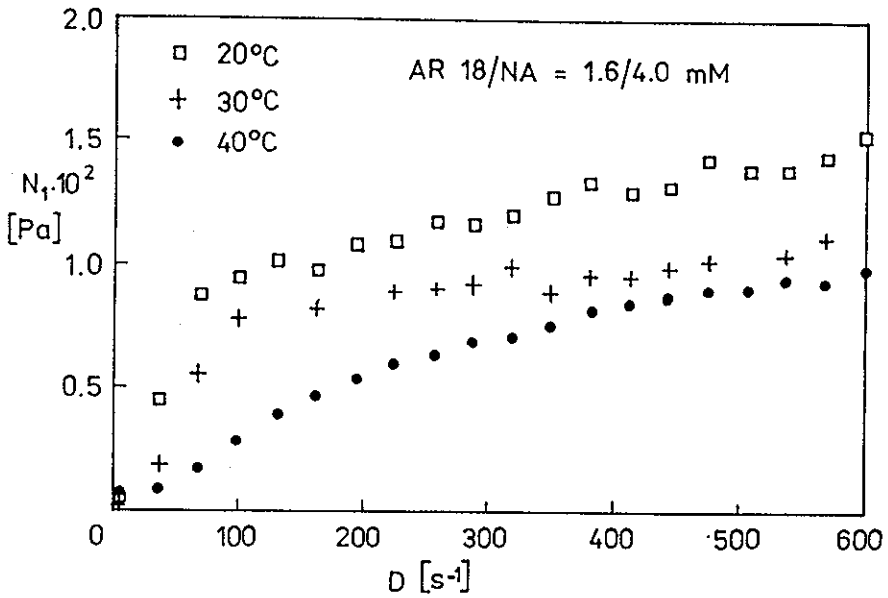


*Figure 4.* Dependence of first normal stress difference on shear rate for AR 18/NA solution with 1.6/4 mM concentration in tap water at 20 and 40°C.

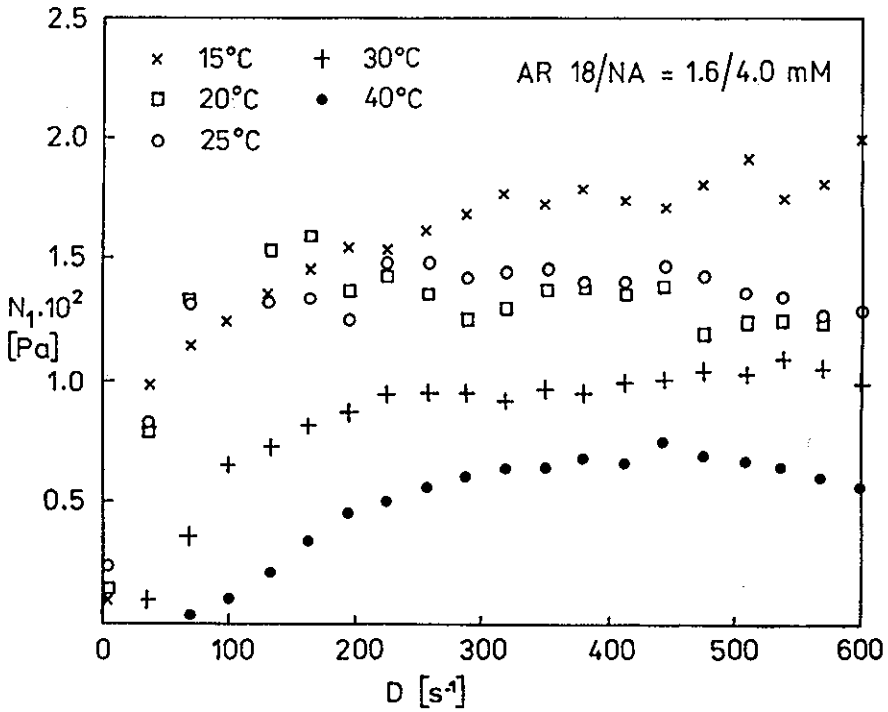


*Figure 5.* Temperature dependence of first normal stress difference on shear rate for AR 18/NA in distilled water. The concentration was 1.6/4 mM.

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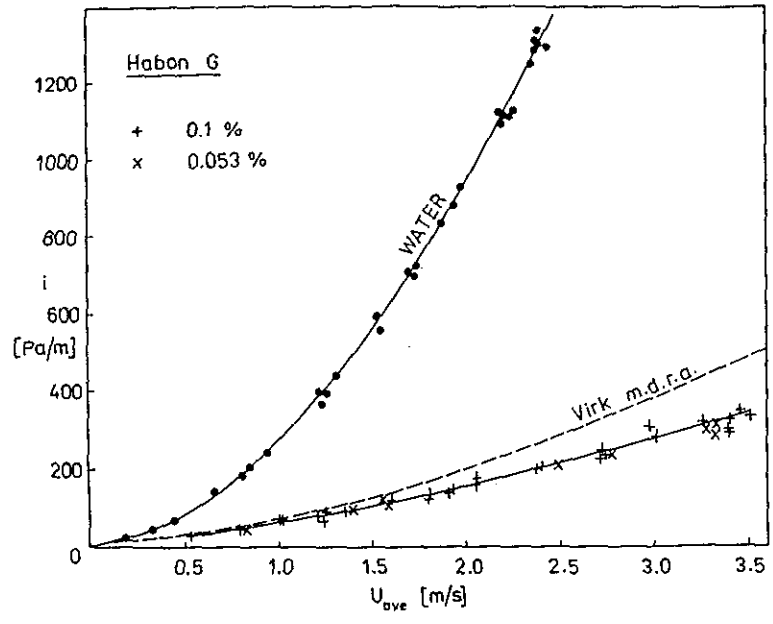


*Figure 6.* Dependence of pressure drop, $i = \Delta P/1$, on average cross section velocity, $U_{ave}$, for Habon G solutions.

in the range studied (0–600 $s^{-1}$). The values of $N_1$ are approximately one half of those for the 5/12.5 mM solution shown in Figure 3. $N_1$ vs. $D$ data for the same surfactant mixture in distilled water are shown in Figure 5. Values of $N_1$ at 20 and 40°C are lower in distilled water than in tap water at high shear rates. $N_1$ decreases as temperature rises for both tap and distilled water from 0 to 40°C. Light scattering data [14] show that the average hydraulic radius of a similar surfactant is larger in tap water than in distilled water which is probably the source of the difference in $N_1$ values. The differences in size and in $N_1$ are surprising as the large excess of sodium chloride and sodium salicylate electrolytes present would be expected to overshadow the electrolytes present in the tap water. Specific ions present in the tap water apparently cause changes in the micelle structure. Research of the water quality on rheological properties of these solutions is underway.

## DRAG REDUCTION

Turbulent flow pressure drop measurements in the 39.4 mm tube system on two other surfactants have been reported previously [6, 15]. At sufficiently high concentrations of Habon G, drag reduction greater than predicted by the Virk maximum drag reduction asymptote [16, 17] was observed. More extensive Habon G data than reported in [6] are presented in Figure 6, where the lower line shows the maximum drag reduction measured in the 39.4 mm ID tube for these surfactants. The plot $i = \Delta p/1$ vs. $U_{ave}$ includes nothing but two directly measured quantities,

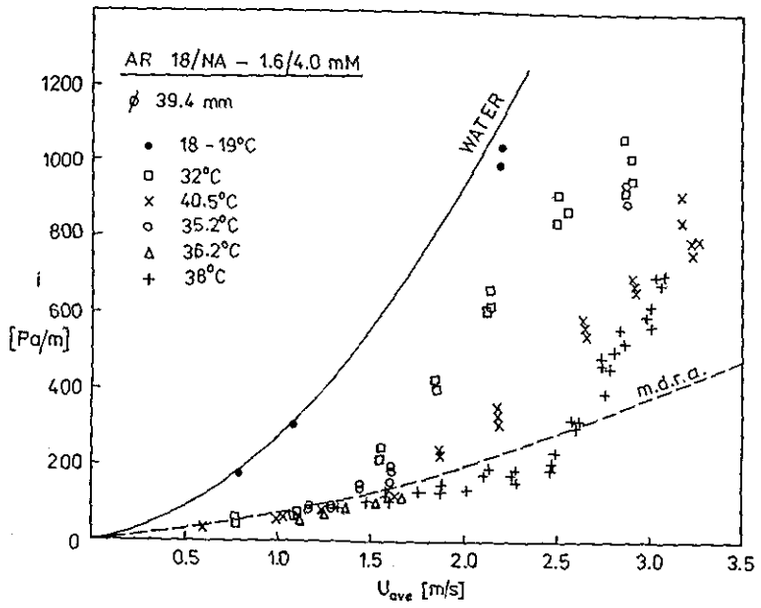


*Figure 7.* Dependence of pressure drop on average velocity for AR 18/NA solution with 1.6/4 mM concentration in 39.4 mm ID tube showing the influence of temperature and time of pumping. Experiments were done in the order listed in the legend.

$\Delta p$ and $U_{ave}$, and no property of the liquid is involved except for the density, and the solution density is the density of water.

Pressure drop data for the 1.6/4.0 mM solution of Ar 18/NA are shown in Figure 7. Measurements were made in the same order listed on the legend, that is the solution at 18–19°C was measured first while that at 38°C was measured last. Thus, the last measurements were made on the solution after it had undergone considerable shear in the previous experiments in the recirculation loop.

Chou [13] reported no drag reduction for the 5/12.5 mM AR 18/NA distilled water solution at 20°C in a 6 mm ID steel tube and significant drag reduction at 30°C and up to 90°C. Data at the lower concentration in tap water shown in Figure 7 show no drag reduction at 18–19°C but significant drag reduction at 30°C and above. Of particular interest is the fact that significant normal stress differences are observed at 15 and 20°C at both concentrations (Figures 3, 4 and 5) but no drag reduction was observed at 20°C by Chou [13] for the higher AR 18/NA concentration or here at 18–19°C for the lower one. Furthermore, in this temperature range (20–40°C) drag reduction increases with temperature while first normal stress difference as well as apparent viscosity decrease. Thus, the existence of significant first normal stress difference is clearly not a determinant of the existence of drag reduction.

This is reminiscent of an opposite effect in flow birefringence data where birefringence was observed in only part of the temperature range in which drag reduction was observed for ET 13/NA at the same molar concentrations [5, 18].

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While drag reduction has been attributed to the viscoelastic nature of high polymer and surfactant solutions, there is no consensus on which viscoelastic characteristic is responsible for the phenomenon. It has been postulated that high extensional viscosity which would inhibit the axial stretching flows involved with near wall eddy propagation is the key factor. Unfortunately, techniques for measuring extensional viscosity in dilute, low viscosity solutions are not yet available to test the hypothesis. What is clear from data, however, is that $N_1$, the first normal stress difference is not the viscoelastic characteristic that causes drag reduction.

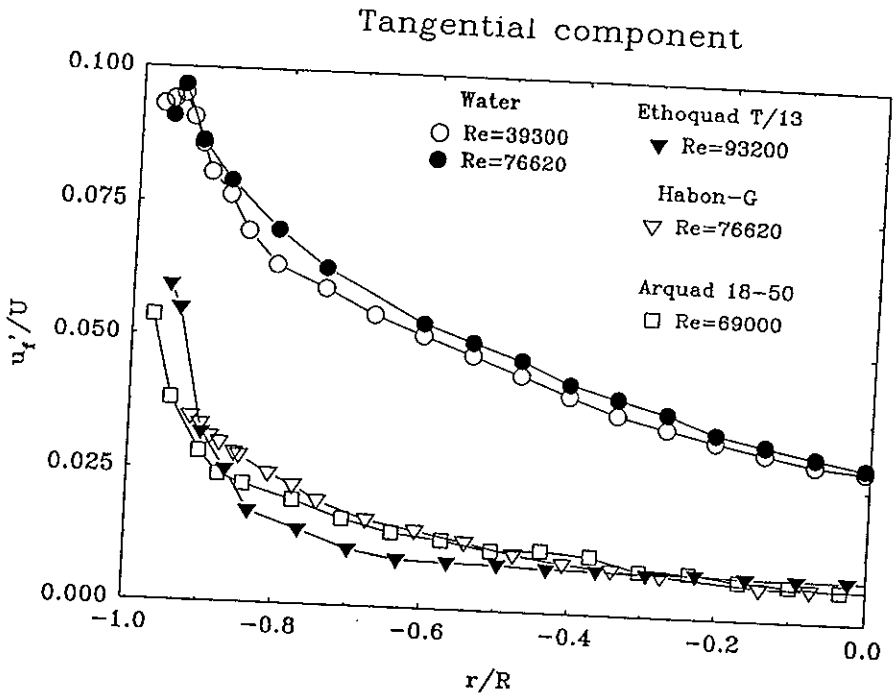
The amount of drag reduction observed increased with time of pumping (amount of shearing) in these experiments, suggesting that measurements at 32, 40.5 and 35.2°C probably would have showed high drag reductions at higher velocities (higher critical wall shear stress for breakdown of structure) if the solution had been sheared (pumped) for a longer period of time before measurements were made at these temperatures. However, we believe that an ultimate efficiency at a certain point will be reached (in the similar way as the ultimate viscosity is approached during pumping in a closed loop which was shown in [10]).

Note that even at the lower 1.6/4.0 mM concentration the 38°C data lie below the maximum drag reduction asymptote for polymers until the critical wall shear stress is exceeded at about 2.5 m/s.

## TURBULENCE INTENSITIES

Turbulence intensity profiles in three directions for Habon G (2 mM), ET 13 (2.5 mM) plus NA (6.25 mM), AR 18 (1.6 mM) plus NA (4 mM) and water are shown in Figures 8a–c. The data are reported as the ratio of root mean square of local velocity component fluctuation to local mean velocity. In approximately the same Reynolds number range, tangential intensities for all these surfactants are lower by one half to two thirds than those of water. Similarly, the radial intensities were reduced to 20–30% of the water values. Longitudinal turbulence intensities are lower than those in water in the central region of the velocity profile.

These measurements were made at a range of concentrations and velocities at which the maximum drag reduction for the Habon G and AR 18/NA solutions was reached and where the maximum drag reduction asymptote for polymers was exceeded. The measurement of turbulence intensities for ET 13/NA solution was made at lower drag reduction levels [15] above the Virk maximum drag reduction asymptote. We can clearly see in Figure 8 that curves of turbulence intensities of ET 13/NA solution rise sharply at a greater distance from the wall than those of AR 18/NA or Habon G. This is most pronounced in the longitudinal turbulence intensities which exceed those of water when $r/R$ exceeds 0.9. The flow regimes with higher levels of drag reduction maintain lower intensities than ET 13/NA close to the wall. Some of the turbulence intensity results for Habon G were reported earlier [6]. Together with new, more extensive measurements, they are shown here for comparison.



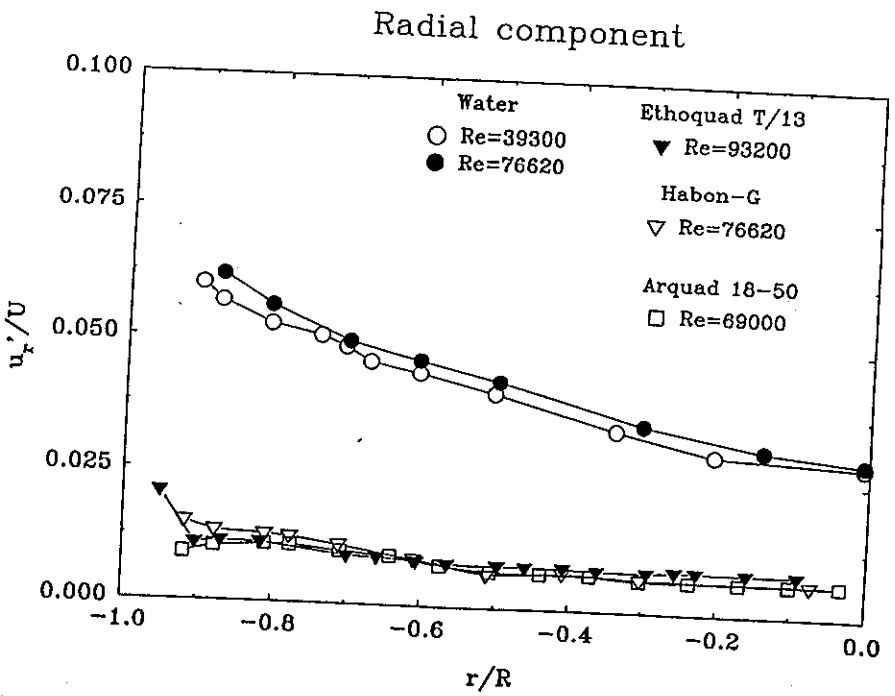




*Figure 8.* Intensities of turbulence in water and in three different surfactant solutions. $U$ is local mean velocity, $u_z'$, $u_r'$ and $u_f'$ are root mean squares (in [m/s]) of longitudinal, radial and tangential velocity fluctuations, respectively, $R$ is the tube radius and $r$ the distance from the tube axis.

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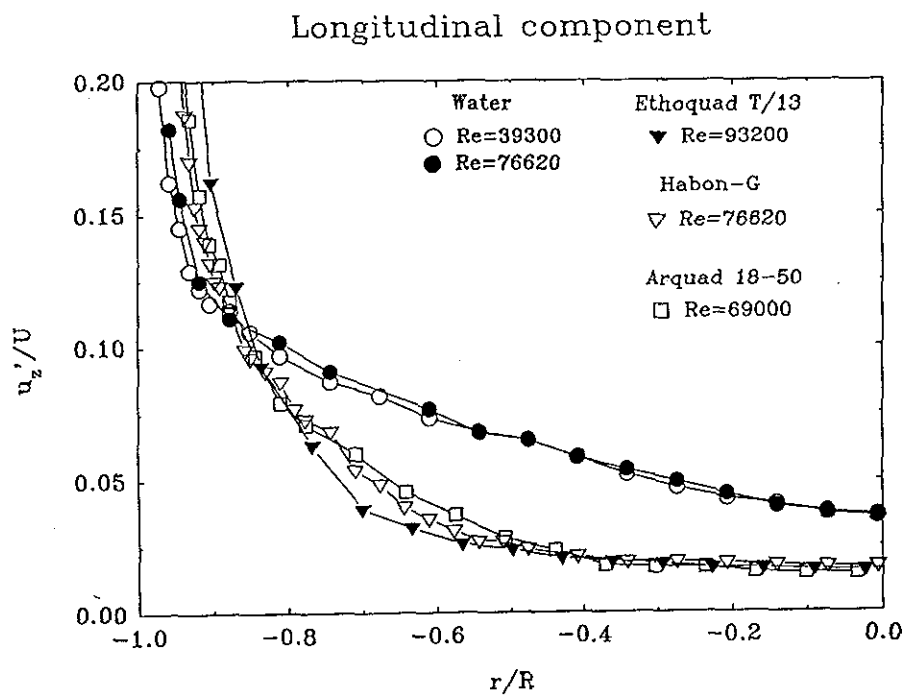


*Figure 8.* (Continued)

Just as the presence of drag reducing surfactant reduces the momentum transfer rate giving lower pressure drops, the surfactants also cause a reduction in heat and mass transfer rates. The small cross fluctuation intensities are therefore not surprising. The presence of the micelles and the microstructures they form, which may impact high extensional viscosities to the solutions and suppress the eddy formation and propagation near the wall, may be the cause of the reduced tangential and radial intensities. They may also reduce axial intensities of turbulence from $r/R = 0$ to approximately 0.85 but, surprisingly, not at the wall. In the wall region, axial turbulence intensities for all surfactant solutions were higher than for water, similar to results in polymer solutions [19]. The values of axial velocity fluctuations in the wall region and thus also the root mean square axial velocity fluctuation, $u'_z$, were generally smaller than in water (see Figure 9) but due to much smaller mean local velocity, $U$, in this region (compared to $U$ in water), the intensity of turbulence, $u'_z/U$, is larger.

Figure 9 shows that longitudinal fluctuations are higher in Habon G solution than in water only at the smallest Reynolds number and at $r/R$ above 0.7. They are also higher with ET 13/NA at $r/R$ above approximately 0.85. The drag reduction of this solution did not reach the Virk maximum drag reduction asymptote. We were not able to obtain data closest to the wall to determine maximum intensities

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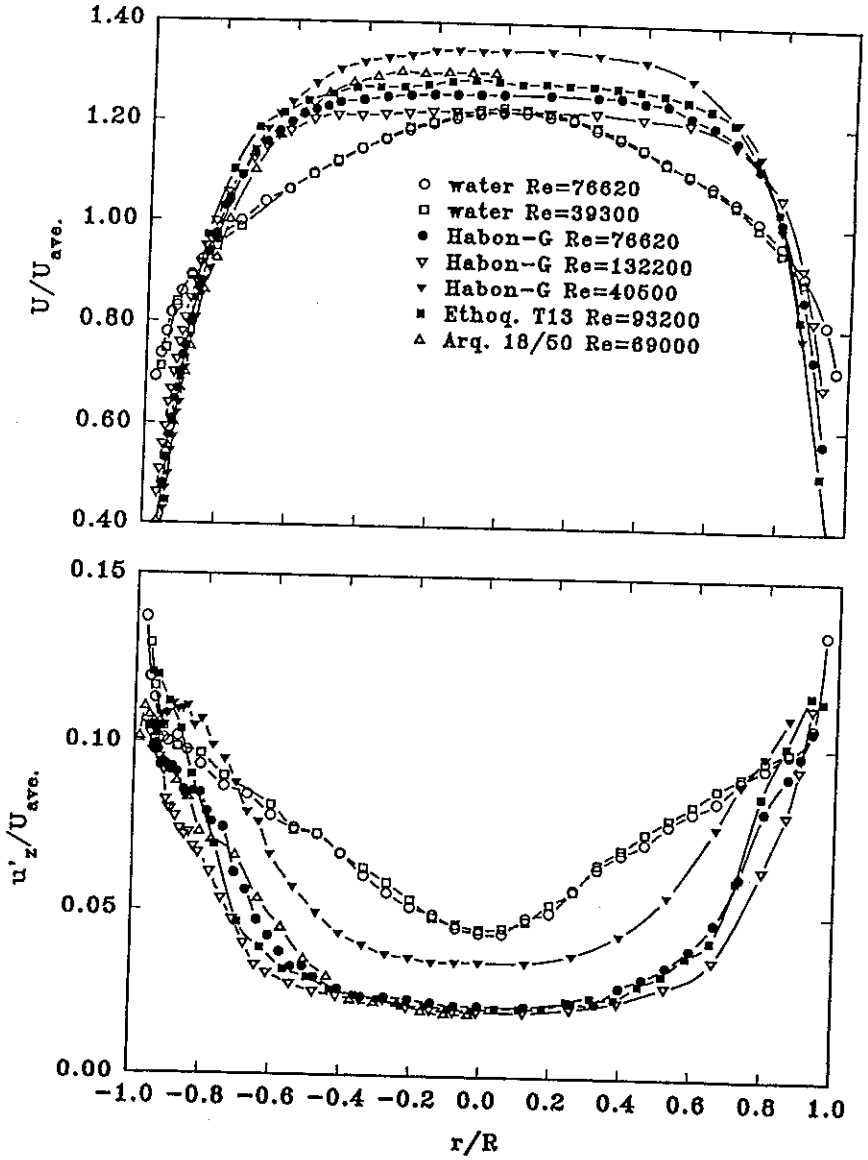


*Figure 9.* Velocity profiles and RMS of longitudinal fluctuations normalized by the same quantity, $U_{ave}$.

and their locations for surfactant solutions. This would be an interesting area for future research.

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## Conclusions

1. The used surfactant solution AR 18/NA shows a high zero-shear viscosity followed by shear thinning and then by a shear-induced structure at high shear rates which is probably caused by alignment of micelle aggregates and is accompanied by increased effectiveness in drag reduction.
2. Drag reduction, exceeding predictions of the Virk maximum drag reduction asymptote, is observed with this cationic surfactant-counterion solution.
3. Significant normal stress differences exist at temperatures where no drag reduction is observed for AR 18/NA solutions indicating that the existence of first normal stress differences does not necessarily lead to drag reduction.
4. The value of the first normal stress difference decreases with temperature in AR 18/NA solutions while drag reduction effectiveness increases with temperature.
5. AR 18/NA in tap water has higher $N_1$ values than in distilled water, which may be due to larger micelles in tap water.
6. Tangential and radial turbulence intensities for drag reducing surfactants are much lower than those for water. Axial (longitudinal) intensities may be higher or lower than those for water depending on the radial position.

## Acknowledgements

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# New Limiting Drag Reduction and Velocity Profile Asymptotes for Nonpolymeric Additives Systems

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Reduction of friction losses in turbulent flows caused by the presence of low concentrations of aluminum disoap and high polymer additives was first reported about fifty years ago (Mysels, 1949; Toms, 1948). Since then there have been a large number of studies of drag reduction, most of which used high polymers, and several excellent reviews exist (Patterson et al., 1969; Hoyt, 1972; Virk, 1975; Sellin, et al. 1982a,b; Hoyt, 1986; Shenoy, 1984). High polymer additives have found important niche applications in crude oil and petroleum product transport (Burger et al., 1982; Motier et al., 1984), in fire fighting (Union Carbide, 1966), in increasing sewer flows (Sellin, 1977) and in jet cutting (Summers and Zakin, 1975), Virk et al. (1970, 1971) proposed equations for a limiting maximum drag reduction asymptote for high polymers and also for an elastic sublayer velocity profile limit. We show here from our experimental data and from those of a number of other investigators (Fankhänel, 1991; Weber, 1990; Althaus, 1991; Ohlendorf and Schwarz, 1984; Pollert et al., 1993; Myska and Vlasak, 1988; Sylvester and Smith, 1979; Myska and Vocel, 1977; McMillan et al., 1971) that both limits proposed by Virk are not valid for aqueous surfactant and aluminum disoap in hydrocarbon systems. New limiting equations for them are proposed. These different behaviors suggest that the mechanism for high polymer drag reduction is probably different than that for micellar systems.

Polymer effectiveness is dependent on the presence of high molecular weight species which limits their applications because of the susceptibility of high molecular weight components to degradation in high shear flows or extensional flows. The former are encountered when passing through pumps and the latter in expansion or contraction flows. Thus, high polymer additives are limited to once-through flows and are not suited for recirculation systems.

In the past decade, considerable interest has developed in nondegrading or "repairable" drag reducing additives for use in district heating and cooling systems to lower the pumping energy requirements (Rose et al., 1984; Pollert et al., 1994). In district heating systems, cogeneration or waste heat sources are used to heat water in a primary loop which circulates the hot water to heat-exchange stations. The heat exchanged to secondary loops provides hot water to heat nearby buildings or to heat hot water for household use. They are widely used in northern and eastern Europe, and their use in the U.S., Canada, Japan, and Korea is expanding. District heating systems conserve energy because of their use of waste heat and centralized production and distribution of heat, and their elimination of often inefficient burners in individual buildings. Thus, they reduce the amount of fossil fuel burned. District cooling systems operate in a similar manner.

Most promising are cationic surfactants of the quaternary ammonium type with appropriate organic counterions. Under the right conditions of surfactant/counterion chemical structures, ratios, concentrations and temperature, they form rod-like micelles. The resulting microstructure imparts viscoelasticity to the solution. The microstructure is mechanically degraded when passing through a high shear pump such as a centrifugal pump, but the structures reform quickly no matter how many times they are broken up by shear. Though surfactant drag reducing additives require higher concentrations than high polymers, their long life and greater potential percent reduction in energy loss make them very attractive for recirculation flows.

Virk et al. (1970, 1971) examined friction factor and velocity profile results for a large number of high polymer solutions, mostly but not all, in water. They noted that at relatively low concentrations many solutions reached lower limiting values in their frictions factor—Reynolds number data. Virk et al. (1970) proposed an equation for the limiting maximum drag reduction asymptote (MDRA). The limiting equation is independent of polymer species, molecular weight, and concentration. The equation, often referred to as the Virk MDRA is

$$\frac{1}{\sqrt{f}} = 19\log\left(N_{Re}\sqrt{f}\right) - 32.4 \qquad (1)$$

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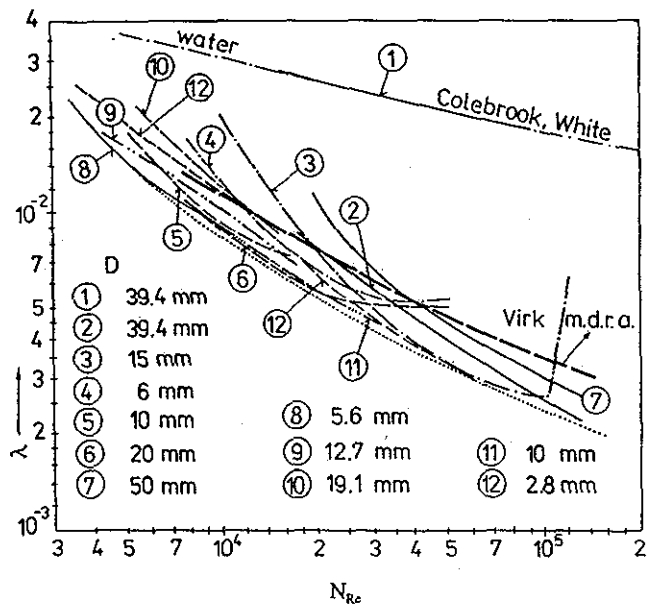


**Figure 1. l vs. *Re*.**

(1) Our data for water; (2) our data for Habon G, Ethoquad T/13/50 (tallow tris-hydroxyethyl ammonium acetate) with sodium salicylate (1.45 mM/3.9 mM) and Arquad 18/50 (octadecyl trimethyl ammonium acetate) with sodium salicylate (1.6 mM/4 mM) in water; (3) data of Fankhänel (1991), Weber (1990), and Althaus (1991) for Habon G, Dobon G and Obon G ($C_{16}H_{33}$, $C_{18}H_{37}$ and $C_{22}H_{45}$ dimethyl hydroxyethyl ammonium-3 hydroxy-2 naphthoate) in water; (4) data of Ohlendorf and Schwarz (1984) for Habon (hexadecyl trimethyl ammonium-3-hydroxy-2 naphthoate) (1,000 ppm) in water; (5) (6) data of Pollert, et al (1993) for Habon G (2 mM) in water; (7) data of Myska and Vlasak (1988) for SEAN (a 1.2 g/L mixture of Carboxy pentadecyl trimethylammonium bromide, commercial septonex and α-naphthol) in water; (8) (9) (10) data of Sylvester and Smith (1979) for aluminum dialkyl phosphate (300 ppm) in kerosene; (11) data of Myska and Vocel (1977) for SEAN (1.55 g/L) in water; (12) data of McMillan et al. (1971) for aluminum distearate (6,000 ppm) in toluene; .....-limiting drag reduction curve for surfactants and aluminum disoaps.

or

$$f \cong 0.58\ N_{Re}^{-0.58}\ (N_{Re}\ 4{,}000-40{,}000) \qquad (2)$$

where $f$ is the Fanning friction factor and $N_{Re}$ is the Reynolds number. A number of investigators using surfactant drag reducing additives and aluminum disoaps have observed friction factors which lie below the Virk MDRA. Data for 1,000 ppm Habon G (hexadecyl dimethyl hydroxyethyl ammonium-2-hydroxy-3-naphthoate), for several other dilute aqueous surfactant solutions, and for two aluminum disoap additives in hydrocarbon solvents are shown in Figure 1. In each of these systems, friction factor data significantly below the Virk MDRA are observed. A limiting curve which envelopes these data is shown in Figure 1. Its equation is

$$\lambda = 4f = 1.26\ N_{Re}^{-0.55} \qquad (3)$$

where the Colebrook White friction factor $\lambda = 4f$.

Virk also noted that the mean velocity profile in turbulent pipe flow underwent changes as concentration of polymer increased. An extended steep elastic sublayer region developed outside the viscous wall region ($y^+ (\geq 11.6)$. Still further from the wall, a core region profile extended parallel to the Newtonian solvent core profile to the center of the pipe. Eventually, at high concentration, the steep region prevails across the entire core (see Virk elastic sublayer in Figure 2): The equation for the elastic sublayer asymptotic velocity profile region for high polymer drag reducing systems is

$$u+ = 26.9 \log_{10} y^+ - 17 \qquad (4)$$

where $u^+$ is mean local velocity divided by friction velocity and $y^+$ is dimensionless distance from the wall. This steep region across the entire profile is associated with the MDRA (Eqs. 1 and 2).

Shown on Figure 2, along with Virk's mean velocity profile asymptote, are $u^+$ vs. $y^+$ data we have obtained for a 500 ppm solution of Habon G at several Reynolds numbers. The velocity profile data away from the wall ($y^+ > 15$) are much steeper than the high polymer asymptote. At high values of $y^+$, which increase with Reynolds number, the profiles bend over and the core region is horizontal. Similar results are obtained with 1,000 ppm Habon G solutions. The limiting equation for the elastic sublayer profile for those data is

$$u^+ = 53.9 \log_{10} y^+ - 65 \qquad (5)$$

The steep velocity profiles of the surfactant solutions are accompanied by friction factors which lie below the MDRA. They indicate a mixing length constant of about 0.02, even smaller than the 0.04 of Eq. 5. Drag reducing surfactant sys-

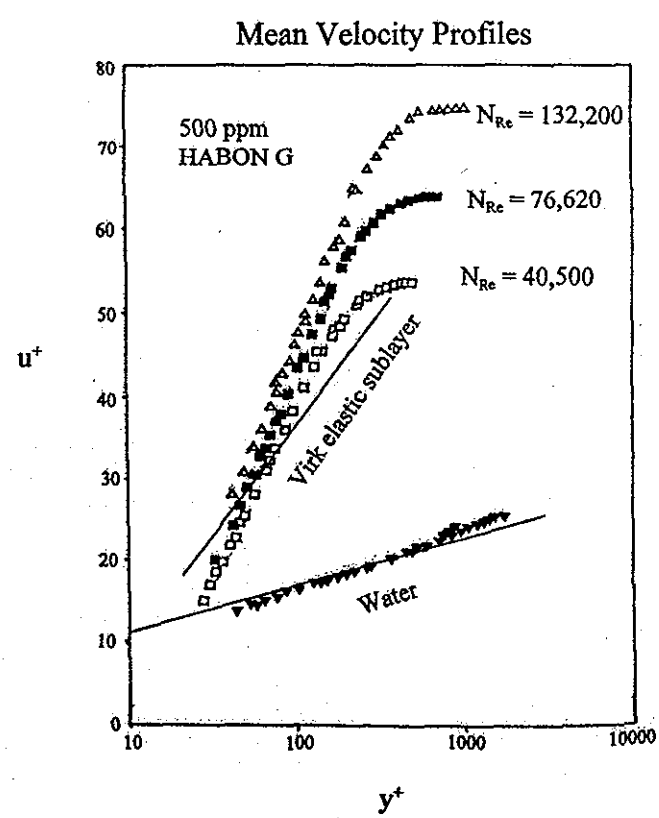


**Figure 2. Mean velocity profiles.**

tem velocity profiles steeper than that predicted by Eq. 5 have also been observed by Bewersdorff and Ohlendorf (1988) and Bewersdorff and Thiel (1993), so 53.9 may not be the ultimate slope.

Thus, there are significant differences in limiting friction factors and velocity profiles between high polymer drag reducing solutions and surfactant and aluminum disoap solutions. This strongly suggests that the mechanisms for drag reduction are different for high polymers and for surfactants and soaps. The reasons for this are not clear, but may be due to network structures formed under mild shear by the rod-shaped micelles in these systems. Cryo-TEM micrographs of cationic surfactant/counterion surfactant systems similar to some of those shown in Figure 1 show tight networks of branched and connected thread-like micelles (for example, See Figure 1, Magid et al., 1990). These have been called shear-induced-structures in which tangled networks of long micellar worms are formed. The network structure must be more effective than even MDRA polymer solutions in reducing turbulence production and turbulent eddy generation.

## Acknowledgment

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J. Hydrol. Hydromech., 45, 1997, 3, 151—172

# MAXIMUM DRAG REDUCTION IN A PIPE FLOW ACHIEVED BY SURFACTANTS

JIŘÍ MYŠKA, ZDENĚK CHÁRA, JACQUES L. ZAKIN

Significant reductions in frictional energy losses or increases in throughput in turbulent pipe flow can be achieved by the addition of small amounts of drag reducing surfactants to the liquids.They are prospective additives for use in recirculation loops of city district heating and cooling systems. Velocity profiles and velocity fluctuations in three directions were measured by means of LDA in a tube of ID 39.4 mm. Low-molecular surfactants showed greater effectiveness in reducing pressure drop than the other well known additives, high-molecular weight polymers. An envelope of maximum drag reduction achieved by surfactants was established using results of other researchers.

KEY WORDS: Turbulence, Drag Reduction, Velocity Profiles, Surfactants.

Jiří Myška, Zdeněk Chára, Jacques L. Zakin: Maximální snížení tření dosažené při proudění micelární povrchově aktivní látky potrubím. Vodohosp. Čas., 45, 1997, 3; 22 lit., 13 obr.

Přídavkem malého množství některé povrchově aktivní micelární látky do kapaliny může být dosaženo významného snížení ztrát při turbulentním proudění v potrubí. Tyto látky snižující třecí odpory jsou zkoumány jako perspektivní aditiva pro použití v uzavřených topných a chladících systémech měst.

Byly měřeny rychlostní profily a fluktuace rychlosti ve třech směrech pomocí LDA v trubici průměru 39,4 mm. Měření ztrát ukázalo, že některé micelární látky mají větší schopnost snížit ztráty než známé polymery. Byla určena obálka největších snížení tření pomocí micelárních látek dosud v odborné literatuře publikovaných.

KLÍČOVÁ SLOVA: snížení tření, rychlostní profil, turbulence, povrchově aktivní látky.

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## Introduction

The phenomenon of active drag reduction by means of an additive was discovered about fifty years ago. Significant reductions in frictional energy losses or increases in throughput in pipe flow can be achieved by addition of small amounts of drag reducing additives in the liquid. Drag reduction by high-molecular weight polymers has received a great deal of attention with hundreds of publications. However, low-molecular weight cationic, anionic and nonionic surfactants have also been found to be good drag reducers and particularly efficient cationic surfactants have been studied intensively in the past few years.

Both kinds of additives have specific advantages and disadvantages. An important advantage of surfactants is their property of immediately regaining drag reduction effectiveness after its loss due to high shear stress soon after the shear has been decreased. A second advantage of surfactants over polymers is their ability to achieve a higher level of drag reduction, although at the cost of higher concentration. A third important property is their effectiveness at high temperatures.

Surfactants have been investigated in our laboratories with the aim of using them in recirculation flows such as city district heating and district cooling systems. The great disadvantage of surfactants is their disposition to create rich foam when free surfaces are exposed to the atmosphere but this is not a problem in closed loops. We have tested a surfactant in a secondary heating system supplying heat to more than 300 apartments in a small town near Prague during the winter 1993–94; pumping energy requirements were reduced by one third when the drag reducing additive was injected into the piping [15]. A detailed description of the test is given in [6].

## Friction coefficient in turbulent flow

The well known equation for turbulent flow of water in smooth pipes is

$$\frac{1}{\sqrt{f}} = 4 \log \mathrm{Re} \sqrt{f} - 0.4, \tag{1}$$

where the Fanning friction coefficient is defined by

$$f = \frac{\Delta p D}{2 \rho l v^2}. \tag{2}$$



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Here the tube diameter is $D$, pressure drop is $i = \Delta p/l$, and $v$ is the cross-sectional average velocity of the liquid with density $\rho$.

Many research results published on drag reducing effectiveness of high polymers agree that there is a maximum pressure reduction that these additives can achieve. Several maximum drag reduction curves either in Prandtl-Karman coordinates $(1/\sqrt{f}, \mathrm{Re}\sqrt{f})$ or simply in coordinates $(f, \mathrm{Re})$ have been proposed. For example Castro and Squire [4] proposed the equation

$$f = 1.4\,\mathrm{Re}^{-0.75}. \tag{3}$$

The next widely recognised equation is the Virk [17] maximum drag reduction asymptote (m d r a)

$$\frac{1}{\sqrt{f}} = 19 \log \mathrm{Re}\sqrt{f} - 32.4 \tag{4}$$

which can be approximated also by the equation

$$f = 0.59\,\mathrm{Re}^{-0.58} \tag{5}$$

for Re = 4,000 to 40,000. All published experiments done with polymers have confirmed that this equation really marks the maximum drag reduction effectiveness in diluted polymer solutions. The slope of the line of the maximum drag reduction asymptote in Prandtl-Karman coordinates – Eq. (4) – is much greater than the slope in Eq. (1) for the flow of water.

## Experimental

Surfactants which are effective drag reducers over a wide range of temperatures, can be used as additives in district heating and district cooling systems. For this purpose some commercial products of Hoechst AG (FRG) and AKZO Chemie (USA) were studied. We have investigated the following surfactants: Habon G, and in a mixture with sodium salicylate, Ethoquad T/13–50 and Arquad 18–50. Further affirmative experiments, though not included in this paper, were done with Ethoquad 0–12 and Arquad 16–50. Their description and formulas were given in [10] and [21].

Pressure drop measurements as well as measurement of turbulent characteristics were made in a 39.4 mm ID glass tube circulation system. The straight test section was 5.8 m long, however, the measuring section was



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about 2 m long. The LDA optics assebled from standard Dantec components were mounted together with Argon-ion laser (Coherent INNOVA 70) on a remote controlled xyz support. A detailed description of the LDA optics and the hydraulic experimental facility can be found in [10].

## Results and discussion

### 1. Velocity profiles

Fig. 1 shows the distribution of the mean local velocity, $u$, normalised by the average cross-section velocity, $v$, vs. dimensionless tube radius, $r/R$, for different Reynolds numbers and different surfactants at optimum concentrations. The velocity profiles of surfactant solutions are flat in the central region which extends from $r/R \cong 0.4$ to $0.7 \cdot u/v$ values are higher than those for water in the core region up to $r/R \cong 0.8$ and they are smaller in the wall region. It should be noted that the ratio $u/v$ (or $u_{\max}/v$) decreases with increasing Reynolds number or with increasing drag reduction efficiency, and that the flat core broadens with increase in Reynolds number. The velocity gradient is very large in the wall region reaching hundreds of reciprocal seconds while in the core region the values are only units of reciprocal seconds. The velocity profile thus strongly resembles the velocity profile of a plastic fluid (e.g. Bingham liquid) with plug flow in the core. This particular shape of the true velocity profile is shown also in the particular shape of the dimensionless velocity profile.

As the flat core broadens with Reynolds number, the velocity ratio $u/v$ in the wall region also increases with Re. The full pattern of such a velocity profile as seen here probably requires an appropriate minimum tube diameter size. A small tube diameter does not give enough space to allow the development of this drag reducing profile. This idea might explain anomalous behavior or very low drag reducing effect often encountered in tubes of small diameters. Such a behavior in 4 and 6 mm tubes was noticed and described e.g. in [14].

Fig. 1 shows the low level of tangential fluctuations in surfactant solution when compared with water, here plotted as root mean square, $u'_f$, normalised by $v$. The same applies for rms radial velocity fluctuation, $u'_r$, in Fig. 2. Only longitudinal fluctuations approach those of water in the wall region, see Fig. 3.

It can be speculated that the large micelle structures are better aligned with the flow direction in the wall region because of the large velocity



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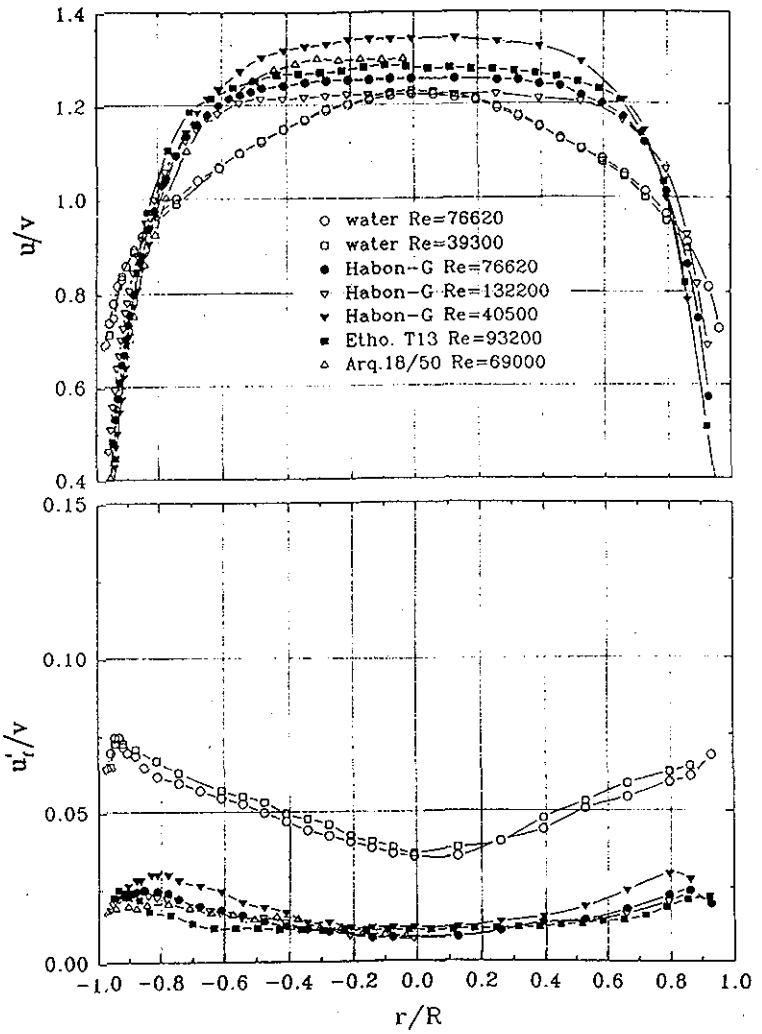


Fig. 1. Velocity and fluctuation profiles in the tube 39.4 mm ID; local mean velocity, $u$, and root mean square tangential velocity fluctuations, $u'_f$, both normalized by mean cross-sectional velocity, $v$, dimensionless distance $r/R$ from the wall.
Obr. 1. Rychlostní profil a profil fluktuací rychlosti v trubici průměru 39,4 mm. Bodová střední rychlost $u$, odmocnina z průměru čtverce tangenciálních fluktuací rychlosti $u'_f$, průměrná průřezová rychlost $v$, bezrozměrná vzdálenost od stěny $r/R$.

gradient. On the other hand, an isotropic micelle structure is expected in the core of the flow with small velocity gradient and thus also a larger effective viscosity than in the wall region may be suggested, too. The



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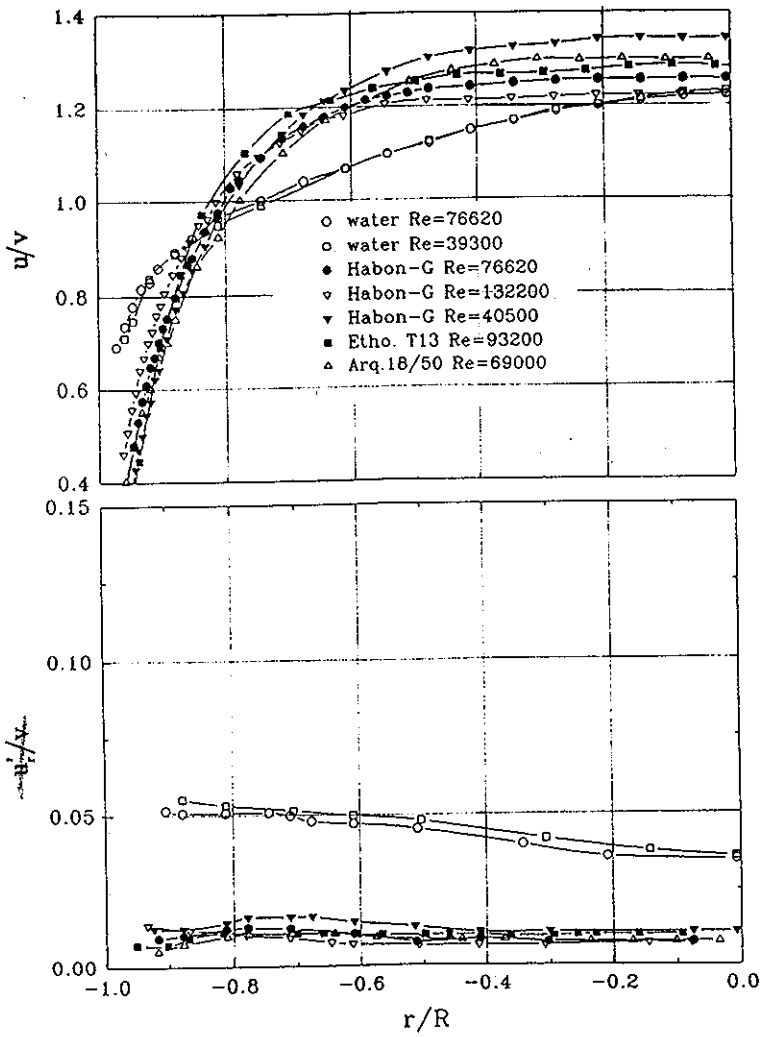


Fig. 2. Velocity and fluctuation profiles; $u/v$, local mean velocity divided by mean cross-sectional velocity, and rms radial velocity fluctuations divided by mean cross-sectional velocity, $u_r'/v$.
Obr. 2. Rychlostní profil a profil fluktuací rychlosti. Bodová střední rychlost $u$, odmocnina z průměru čtverce radiálních fluktuací rychlosti $u_r'$, průměrná průřezová rychlost $v$.

micelle alignment with the flow direction with a large shear rate has been proved by many birefringence measurements and also viscosity curves show minima at large shear rates. Let us consider the micelle structures in a unit volume of the liquid in the wall region. Their projection into the radial



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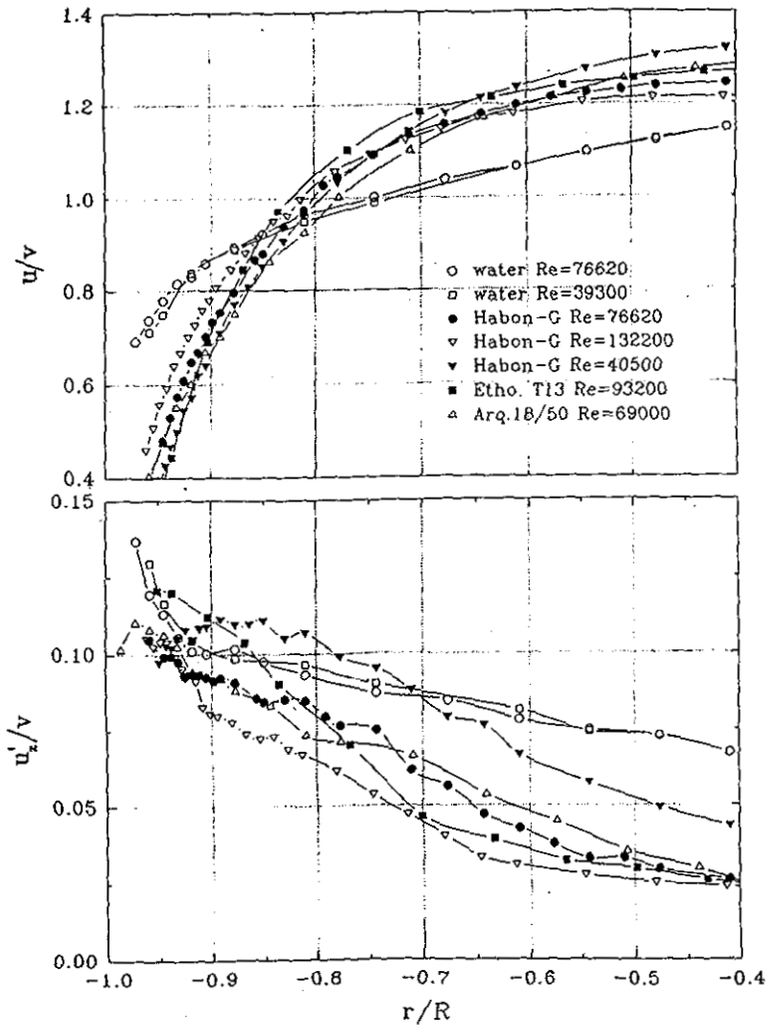


Fig. 3. Velocity and fluctuation profiles: $u/v$ and $u'_z/v$, rms longitudinal velocity fluctuations/mean cross-sectional velocity.
Obr. 3. Rychlostní profil a profil fluktuací rychlosti. Bodová střední rychlost $u$, odmocnina z průměru čtverce podélných fluktuací rychlosti $u'_z$, průměrná průřezová rychlost $v$.

or tangential direction is much greater than their projection into the axial direction. This means greater interference with both radial and tangential fluctuations which are thus suppressed while longitudinal fluctuations are allowed to grow to the water values in the vicinity of the wall as seen in Figs. 1, 2 and 3.



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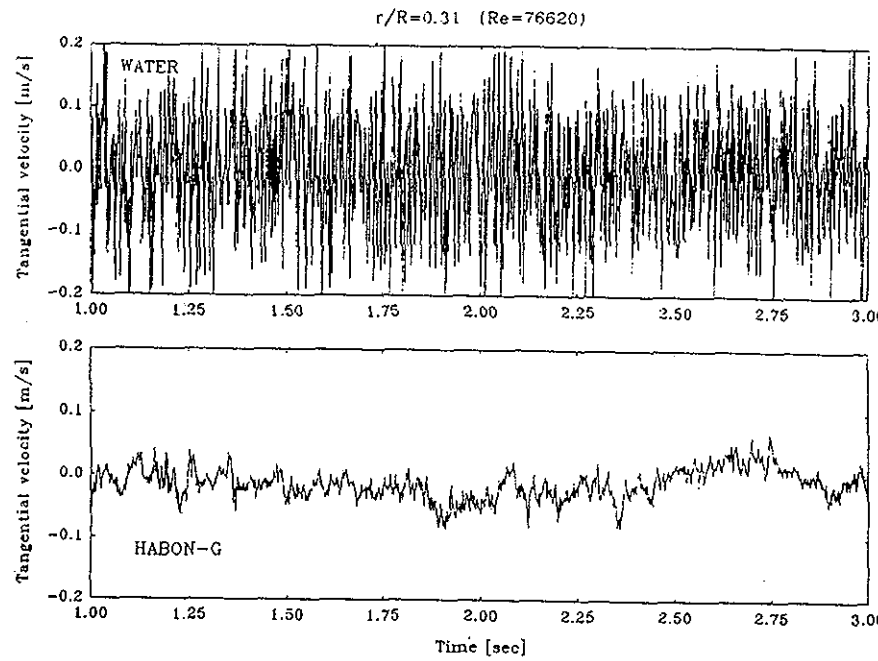


Fig. 4. The time sequence of tangential velocity fluctuations 6.1 mm from the wall. Difference between water without and with the additive at the same Reynolds number.
Obr. 4. Sled tangenciálních fluktuací rychlosti 6.1 mm od stěny ve vodě a ve vodě s povrchově aktivní látkou (PAL) při stejném Reynoldsově čísle.

The time sequences of tangential, longitudinal and radial fluctuations are shown in Figs. 4 to 7. The reduction of both the frequences and amplitudes by addition of a surfactant is clear, eventhough they increase with the approach to the wall.

The dimensionless velocity profiles $u^+$ vs $y^+$ in surfactants differ from those in polymers. For water in the logarithmic layer is

$$u^+ = 5.75 \log y^+ + 5.5 = 2.5 \ln y^+ + 5.5 \quad \text{for} \quad u^+ \geq 11.5 \tag{6a}$$

and in the wall layer $u^+ = y^+$ for $u^+ \leq 11.5$.

Spalding [16] deduces the theoretical equation for both layers along the whole tube radius in the form

$$y^+ = u^+ + 0.1108\,(e^{0.4u^+} - 1 - 0.4u^+) \tag{6b}$$

which he further corrects in order to adjust to Laufer's [7] experimental



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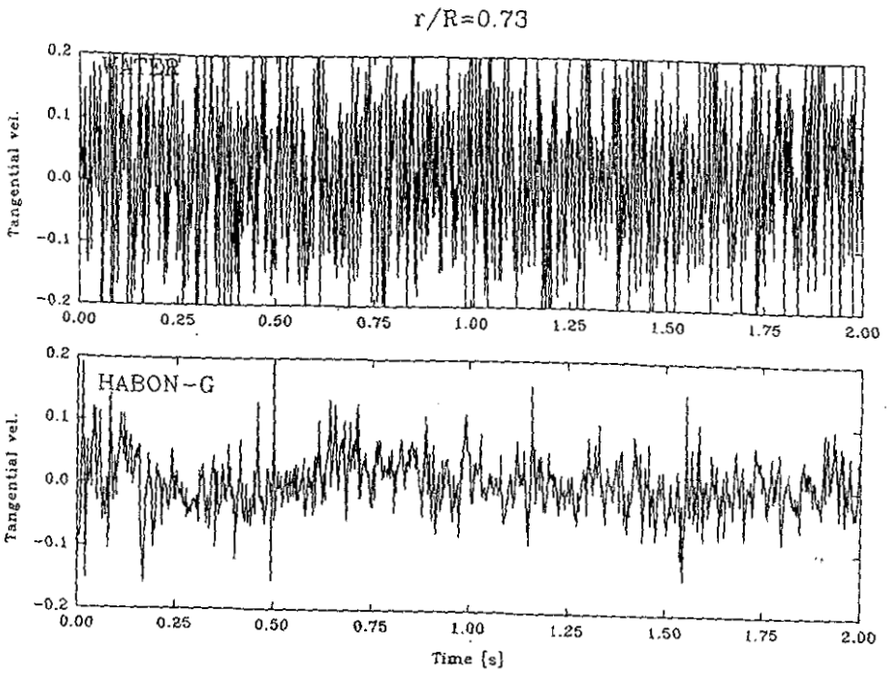


Fig. 5. The time sequence of tangential velocity fluctuations 14.4 mm from the wall. Difference between water with and without additive at the same Reynolds number.
Obr. 5. Sled tangenciálních fluktuací rychlosti 14.4 mm od stěny ve vodě a ve vodě s povrchově aktivní látkou (PAL) při stejném Reynoldsově čísle.

data into

$$y^+ = u^+ + 0.1108\left(e^{0.4u^+} - 1 - 0.4u^+ - \frac{(0.4u^+)^2}{2} - \frac{(0.4u^+)^3}{6}\right). \quad (6c)$$

The ultimate profile deduced by Virk, Mickley and Smith [18] for polymer flow which corresponds to the maximum drag reduction asymptote is

$$u^+ = 11.7 \ln y^+ - 17. \quad (7)$$

In these equations

$$u^+ = u/v_*, \quad y^+ = yu^+/\nu, \quad v_* = \sqrt{\tau_w/\rho}, \quad \tau_w = \Delta p D/4l, \quad (8)$$

where $y$ is the distance from the wall and $\nu$ is kinematic viscosity of the liquid with density $\rho$. Usually the solvent viscosity is considered.



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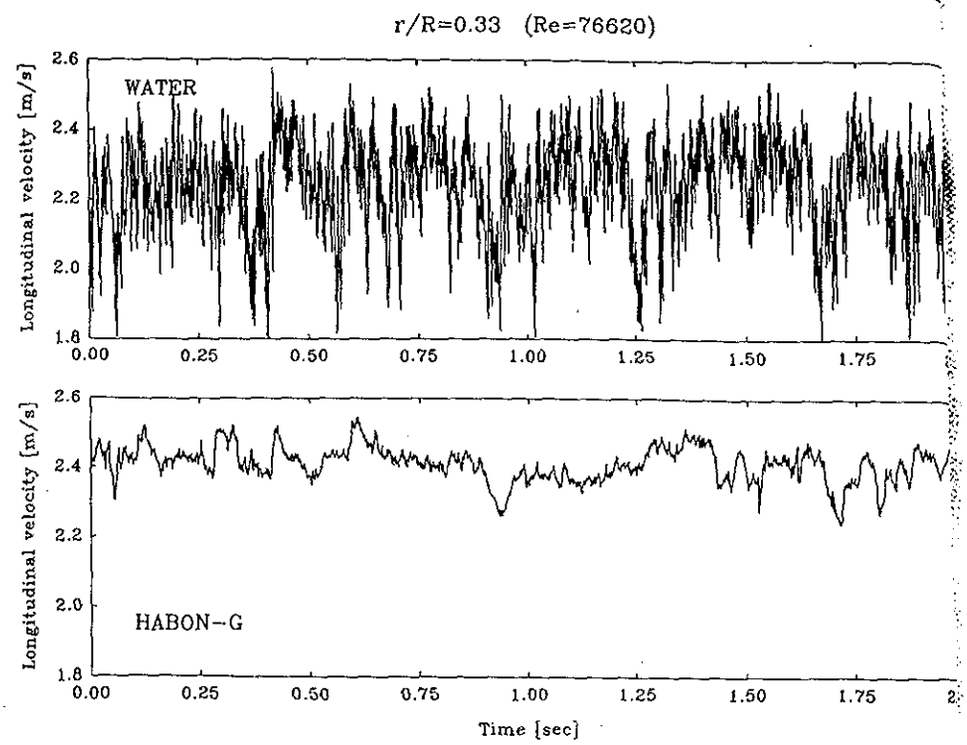


Fig. 6. The time squence of longitudinal velocity fluctuations in water and in the surfactant.
Obr. 6. Sled podélných fluktuací rychlosti.

Fig. 8 shows two mean velocity profiles in surfactants. The data for water lie in a narrow bend following Eq. (6b), which lies below Eq. (6c) in the region of smaller values of dimensionless distance. The measured points for water at higher values of $y^+$ lie above the line defined by Eq. (6a) which, however, is in agreement with published data by many authors. The mean profile in Habon G was obtained with 2 mM concentration of the additive, the concentration at which maximum drag reduction is obtained in a large range of Re. The slope of this line is significantly steeper than that of Eq. (7). It was shown in [22] that mean velocity profiles depend on Reynolds number but the slopes do not differ much. The second mean velocity profile in Fig. 8 was measured for a solution of 2.5 mM Ethoquad T/13-50 with 6.25 mM sodium salicylate, a concentration with relatively low efficiency. Nevertheless, the steepness of the slope is similar to that obtained at optimum conditions with Habon G and it is also greater than



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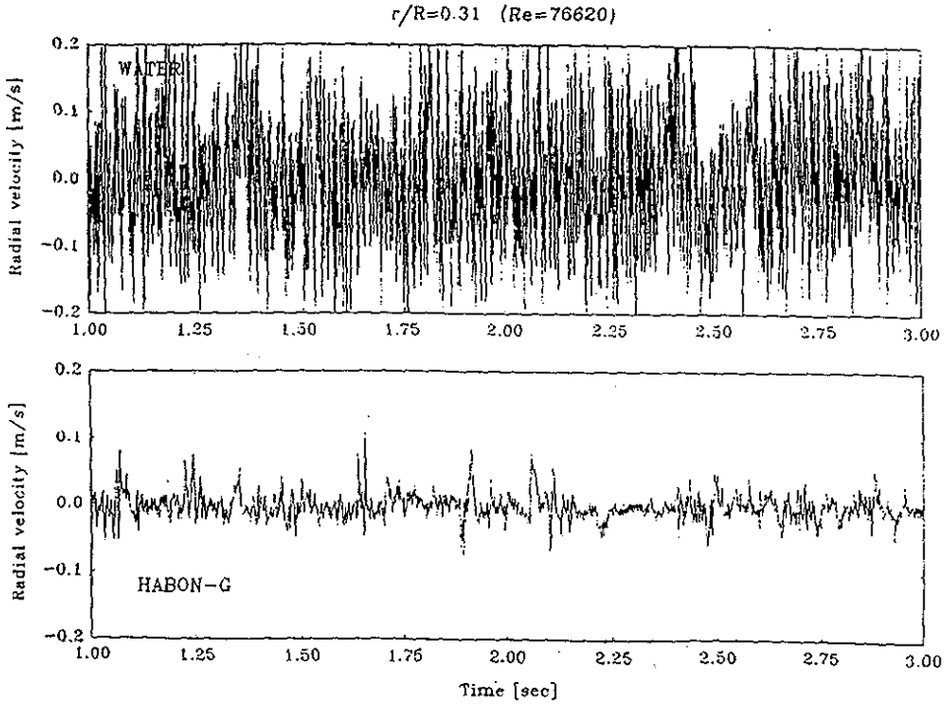


Fig. 7. The time sequence of radial velocity fluctuations in water and in the surfactant.
Obr. 7. Sled radiálních fluktuací rychlosti.

that of Eq. (7). However, this complete velocity profile is positioned below the ultimate profile corresponding to the Virk maximum drag reducing asymptote. Thus the plot of the function $u^+(y^+)$ depends also on the efficiency and concentration of the surfactant.

There is a small uncertainty in the position of the light beam on tube wall. Because of a certain thickness of the beam, its centre can be shifted from the wall by an order of several hundredths of milimeter and therefore our mean velocity profile can be shifted in $\pm y^+$ direction. Furthermore, the solvent viscosity was used in $y^+$. With the true solution viscosity either constant or variable across the diameter the velocity profile would be shifted to the left.

We can compare the mean turbulent velocity profile in surfactants to the schematic three zone polymeric profile published by Virk, Mickley and Smith [18] which they name 1. viscous sublayer, 2. elastic sublayer or interactive zone and 3. Newtonian plug. We recorded the second and third



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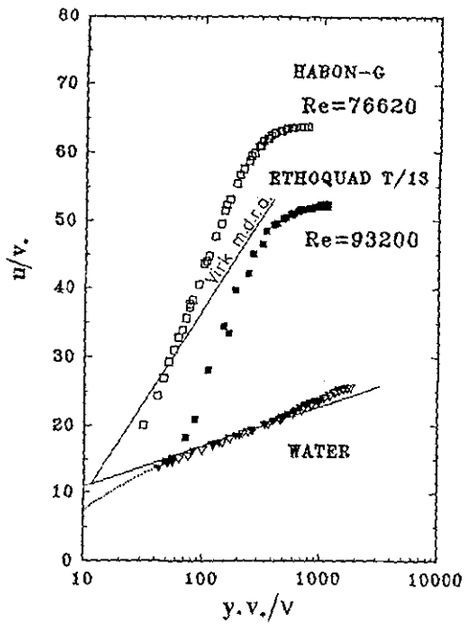


Fig. 8. Dimensionless velocity profiles $u^+ - y^+$ in surfactant solutions: in Habon G at maximum drag reducing effectiveness, in low concentrated Ethoquad T/13-50 with sodium salicylate at small effectiveness. Full line for water: Eq. (6a); dotted line: Eq. (6b).

Obr. 8. Bezrozměrné profily rychlosti $u^+ - y^+$ v roztoku PAL: při maximální účinnosti s Habonem G a při malé účinnosti s málo koncentrovaným roztokem Ethoquadu T-13/50 se salicylanem sodným. Voda: plná čára rov. (6a) a tečkovaná čára rov. (6b).

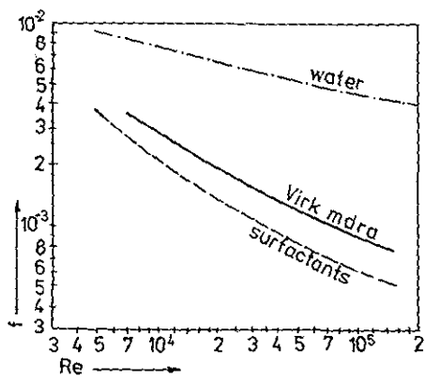


Fig. 9. Friction coefficient $f$ against Reynolds number. Dashed line is the envelope of assembled data with surfactants and aluminum disoaps exceeding Virk maximm drag reduction asymptote as published in [22].

Obr. 9. Součinitel tření $f$ v závislosti na Re. Čárkovaně je vyznačena obálka shromážděných výsledků s PAL podle autorů, uvedených v [22], které leží pod Virkovou asymptotou.

zone in surfactants in our measurements. The second zone must include a transition from the viscous sublayer to a straight line which may have a much steeper slope than the elastic sublayer in polymers. The third zone in surfactants includes a horizontal niveau while the $u^+(y^+)$ curve in polymers in the third zone is usually drawn with a branch parallel (with a vertical shift of $\Delta B$) to the line for water. The horizontal branch of the surfactant



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dimensionless velocity profile, which represents the core region of the flow, is clearly seen in all our measurements [22].

The schematic mean velocity profile in surfactants can thus be described as follows: viscous sublayer

$$u^+ = y^+ \qquad \text{for} \quad y^+ \leq 11.5, \tag{9a}$$

the main logarithmic part of the interactive zone

$$u^+ = G \ln y^+ + F \qquad \text{for} \quad y_1^+ \leq y^+ \leq y_2^+, \tag{9b}$$

the horizontal branch for the core region

$$u^+ = E \qquad \text{for} \quad y^+ > y_2^+. \tag{9c}$$

Approximate values of parameters in Eqs.(9) as calculated from results presented in [22] are in the following table:

| Re | $G$ | $F$ | $E$ |
|---|---|---|---|
| maximum drag reduction (optimum concentration) | | | |
| 40500 | 18.8 | −47.5 | 53.5 |
| 76620 | 20.3 | −51 | 63.5 |
| 132200 | 23.4 | −65 | 74.5 |
| less than maximum drag reduction (from Fig. 8) | | | |
| 93200 | 18.7 | −60 | 52.5 |

This table clearly shows the role of Reynolds number and greater slope in the interactive zone than in Eq. (7). Even higher values of $G$ can be expected. Steeper slopes situated above the ultimate profile in polymers were also measured in a fiber suspension by McComb and Chan [8] and in a surfactant solution by Bewersdorff and Thiel [2].

## 2. Drag reduction effectiveness

We have shown by our measurements in the 39.4 mm pipe [10] and by collection of data published by other authors that some commercial drag reducing cationic surfactants and aluminum disoaps are more effective additives than polymers. This result was discussed in [10, 22] and it is illustrated here by the simplified $f$-Re plot in Fig. 9. The envelope of all assembled data (the authors are named in [22]) with best efficiency is

depicted in the figure by dashed curve. The dynamic viscosity $\mu$ in the definition of Reynolds number

$$\mathrm{Re} = \frac{\rho v\, D}{\mu} \tag{10}$$

is the solvent viscosity at the test temperature. In studies of friction behaviour of drag reducing solutions it is convenient to use the solvent viscosity [17]. It permits comparison of different drag reducing data and comparing measurements with the empirical friction law of maximum drag reduction, Eq. (5). But it is necessary to bear in mind that the $f$-Re plot is unique for every tube diameter.

The limiting curve which envelopes these data is approximately

$$f \approx 0.32\mathrm{Re}^{-0.55} \tag{11}$$

for Reynolds numbers between 4,000 and 40,000.

An optimum concentration exists for each surfactant resulting in maximum effectiveness. Further increase in concentration beyond the optimum does not reduce the friction coefficient. Thus the position of the function $f(\mathrm{Re})$ is not affected since the necessary optimum concentration is reached, but, in combination with the tube diameter, the concentration does affect the value of average velocity in the tube or Re above which maximum drag reduction effectiveness no longer is achieved.

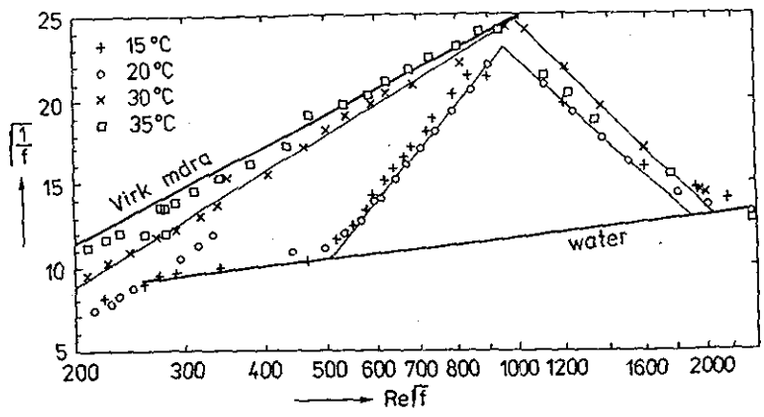


Fig. 10. Recalculated data of Bewersdorff and Ohlendorf [3] and replotted in Prandtl-Karman coordinates.
Obr. 10. Výsledky Bewersdorffa a Ohlendorfa [3], přepočtené do Prandtlových-Kármánových souřadnic.



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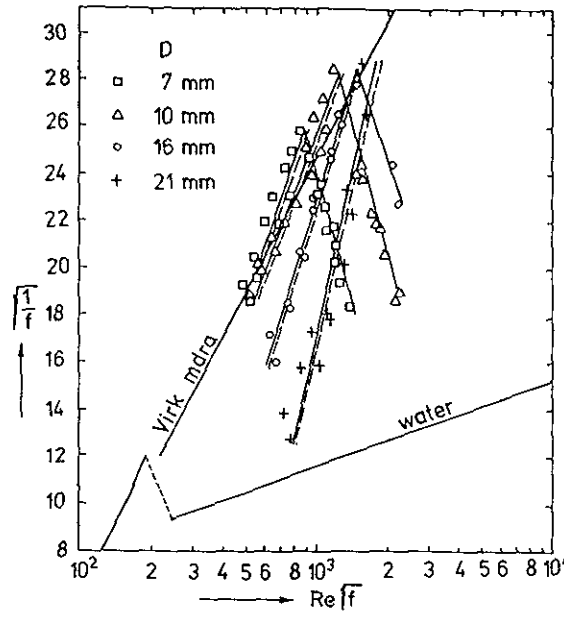


Fig. 11. Drag reduction of 0.0155 wt% carboetoxy pentadecyl trimethyl ammonium bromide with α-naphtol (SEAN) solution in Prandtl-Karman coordinates in tubes of different diameters. Full lines: Re calculated with the viscosity of the surfactant solution, dashed lines: Re calculated with the solvent (water) viscosity.
Obr. 11. Snížení tření v 0,0155-procentním roztoku Septonexu s l-naftolem (SEAN) v P-K souřadnicích v trubicích různých průměrů. Plná čára: Re vypočítané s viskozitou roztoku, čárovaně: Re s viskozitou vody.

Another view of drag reduction effectiveness can be seen using Prandtl-Karman semilogarithmic co-ordinates. Here, the behaviour of surfactants can be interpreted by means of "isosceles triangles". To show this, we have replotted the data of Bewersdorff and Ohlendorf [3] in Fig. 10, although the maximum efficiency in their measurement does not exceed the Virk m d r a.

Earlier an extensive research of drag reduction in suspensions with the aid of another surfactant additive was described in [11, 12, 19]. The flow of moderately concentrated suspensions of small particles (spheres) was investigated in pipes of 7,10,16 and 21 mm ID and the friction of the suspension was decreased by the two-component surfactant SEAN which consisted of carboxy pentadecyl trimethylammonium bromide – commercial name Septonex (SE)-with α-naphtol (AN) with a weight ratio 2.8:1. The friction was influenced by both the suspension concentration (drag increase)



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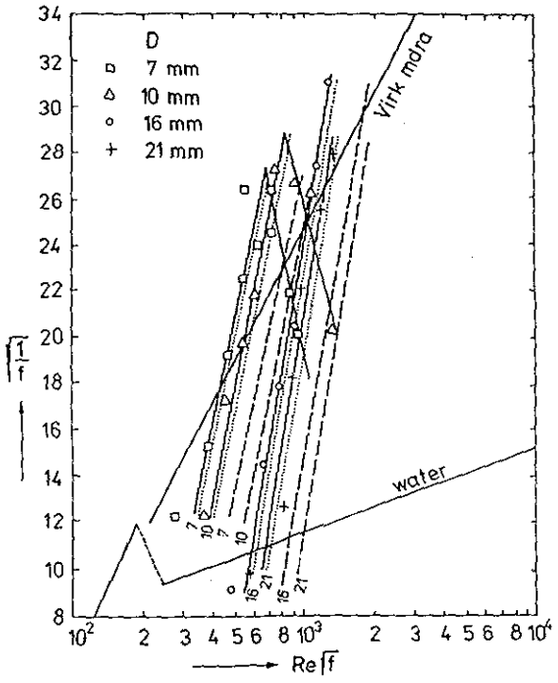


Fig. 12. Drag reduction of 0.0265 wt% SEAN solution in a suspension of 6% volume concentration of rigid particles. Full lines: Re calculated with the viscosity of the suspension with surfactant, dotted lines: Re calculated with the solvent (suspension) viscosity and dashed lines: Re calculated with water viscosity only.
Obr. 12. Snížení tření v 0,0265-procentním roztoku SEAN v suspenzi s objemovou koncentrací tuhých částic 6%. Plná čára – ve výpočtu použita viskozita suspenze s PAL, tečkovaně – viskozita suspenze a čárkovaně – výpočet Re pouze s hodnotou viskozity vody.

and surfactant concentration (drag decrease). The result with 0.0155 wt% of surfactant in tap water is shown in Fig. 11 where the typical "triangles" can be seen. They shift to higher $\mathrm{Re}\sqrt{f}$ values with increasing diameter. Viscosity used in the original paper [19] was the Newtonian viscosity of the surfactant solution (points and full lines). The dashed lines show the recalculation with the solvent (water) viscosity. This figure confirms that drag reduction with this micellar surfactant also exceeds Virk m d r a at a concentration which was not the most efficient one. Similar results are shown in Fig. 12 with 0.0265 wt% surfactant concentration but in this case in a suspension with 6% volume concentration of rigid particles. Again the points and full lines show measured data and Re originally calculated

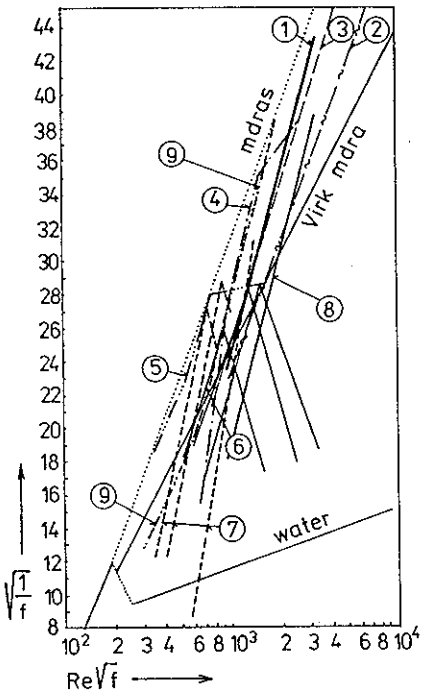


Fig. 13. Some collected data on drag reduction of surfactants in water in P-K coordinates. 1 – our measurement in 39.4 mm tube, 2 – correlation proposed by Althaus [1] for 15 mm tube, 3 – the envelope of Althaus's data, 4 – data by Weber [20] in 15 mm tube, 5 – data by Pollert, Komrzy, Voženílek and Zakin [14] in 20 mm tube, 6 – (thin full lines) data by Myska and Vocel [11] in 7, 10 and 16 mm tubes (drag reduction by surfactant in a suspension) and 7 – (dashed lines) data by Myska [12] in 7, 10 and 16 mm tubes, 8 – (full line) data by Myska and Vlasak [13] in 50 mm tube, 9 – (dash-dash, dot-dot) data by McMillan, Hershey and Baxter [9] in 2.8 mm tube.

Obr. 13. Maximální snížení tření podle různých autorů. 1 – vlastní měření v trubici průměru 39.4 mm, 2 – korelace podle Althause [1] pro 15 mm trubici, 3 – obálka naměřených výsledků podle Althause, 4 – data v 15 mm trubici podle Webera [20], 5 – měření Pollerta a kol. [14] ve 20 mm trubici, 6 – (tence plně) měření Myšky a Vocela [11] v 7, 10 a 16mm trubicích (PAL v suspenzi), 7 – (čárkovaně) data podle Myšky [12] v 7, 10 a 16 mm trubicích, 8 – (plně) výsledky Myšky a Vlasáka [13] v 50mm trubici a 9 – (čárka-čárka, tečka-tečka) výsledek podle McMillana a kol. [9] ve 2.8mm trubici.

with the apparent viscosity of the suspension plus additive. If we replot the result using solvent viscosity (in this case viscosity of the suspension only), we obtain the dotted lines. These lines clearly show that surfactant yields lower friction factors than polymers. The influence of the pipe diameter is also shown in Figs. 11 and 12.

Data of different authors are collected in Fig. 13. Line No. 1 comprises our own measurements with different surfactants (Habon G, Ethoquad T/13–50, Arquad 18–50) in water at different concentrations and different temperatures. Fankhänel [5] and Althaus [1] published measurements with Habon G, Obon G, Dobon G, Obon and Dobon (all products of Hoechst AG) in a tube of 15mm ID. A correlation was proposed in the



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form

$$\sqrt{\frac{1}{f}} = 26.6 \log \mathrm{Re}\sqrt{f} - 57. \tag{12}$$

This equation is drawn in Fig. 13 as Line No. 2. Line No. 3 marks measured points with greatest divergence from No. 2. Weber's [20] data for Habon G at 60°C were originally plotted in $\lambda$ – Re diagram ($\lambda = 4f$) and in Fig. 13 have been replotted. They are represented by Line No. 4. Pollert, Komrzy, Vozenilek and Zakin [14] measured friction coefficients in solutions of Habon G and Ethoquad T/13–50 with sodium salicylate. They found the most effective drag reduction in the 20 mm tube and these data are redrawn as Line No. 5. Line No. 6 shows results from experiments with surfactant SEAN (from Fig. 11) in water and Line No. 7 shows the same surfactant with suspension of 6% rigid particles (from Fig. 12). The tube diameters in these experiments were 7, 10 and 16 mm [11, 12]. The solvent Reynolds number is also used in Fig. 13. Line No. 8 shows recalculated data from the 50 mm tube for SEAN [13] and No. 9 is 2.8 mm tube data obtained with aluminum distearate by McMillan, Hershey and Baxter [9]. This line as well as Line No. 7 extends to the dotted line which marks the maximum limit for surfactants and aluminum disoaps.

An approximate maximum drag reduction in surfactants (m d r a s) can be drawn as the envelope to these curves with an equation

$$\sqrt{\frac{1}{f}} = 26 \log \mathrm{Re}\sqrt{f} - 48. \tag{13}$$

This line is situated above the line defined by Eq. (4) as seen in Fig. 13.

We believe that more data showing smaller friction factors achieved in surfactants will be found and approximate constants in Eq. (13) will be verified.

The proposed m d r a s does not describe real relation between friction coefficient and Reynolds number in a single pipe but describes the limit of maximum efficiency of drag reduction which can be reached in pipes of any diameter by a surfactant. It is an envelope reflecting the best results reported by different authors thus far.

It was possible to integrate the mean velocity profile and compare the constants of logarithmic members in $u^+(y^+)$ equation and $f(\mathrm{Re}\sqrt{f})$ equation for polymer flows. However, as our Eq. (13) is not an equation



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describing true flow conditions in the tube, and as the mean velocity profile of surfactant solution is dependent on Reynolds number, there does not exist its vis-a-vis counterpart in $u^+(y^+)$ function with quantities $G$ and $F$.

## Conclusion

The turbulent velocity profiles of surfactant solutions differ from those of polymer solutions. As in polymers, three zones of the dimensionless velocity profile can be distinguished. However, the dimensionless velocity profile in a surfactant depends on Reynolds number, the slope in the interactive zone is steeper than for polymer solution flow. The main part of the third zone, the plug zone, is horizontal.

Surfactants show greater ability to decrease friction coefficients than polymers. Available data from the literature and our own experiments permit us to write the equation for a m d r a s, based on an envelope covering results with maximum drag reduction effects achieved with surfactants.

Velocity fluctuations in radial and tangential directions are suppressed by micelles in the whole tube diameter, while the longitudinal velocity fluctuations are smaller only in the central part of the tube where the apparent viscosity of the surfactant solution is high. In the wall region, the longitudinal fluctuations are close to the fluctuations in water. The decrease of both frequency and amplitude of the fluctuations has been shown.

Excess surfactant concentration over the optimum concentration does not increase the maximum drag reduction in a given tube diameter and at a certain temperature. However, the friction factor – Reynolds number results are markedly dependent on the tube diameter; the concentration can affect the range of Re (at an unchanged temperature) with maximum drag reduction.

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# MAXIMÁLNÍ SNÍŽENÍ TŘENÍ DOSAŽENÉ PŘI PROUDĚNÍ MICELÁRNÍ POVRCHOVĚ AKTIVNÍ LÁTKY POTRUBÍM

Jiří Myška, Zdeněk Chára, Jacques L. Zakin

Ztráty třením při turbulentním proudění v potrubí mohou byt významně zmenšeny přidáním malého množství některé micelární povrchově aktivní látky (PAL). Perspektiva použití těchto aditiv v městských otopných či chladících systémech vyvolala zájem o jejich všestranný výzkum. Důkaz o možnosti použití v uzavřeném sekundárním otopném systému byl podán provozním pokusem na sídlišti Kladno-Kročehlavy, kde bylo teplem zásobováno více než 300 bytů. Elektrický příkon na čerpání teplé vody mohl být trvale snížen o celou jednu třetinu při koncentraci aditiva již pod 0,05%.

V článku jsou uvedeny výsledky měření s LDA na trubici profilu 39,4 mm. Fluktuace rychlostí v radiálním a tangenciálním směru jsou přítomností micel potlačeny v celém profilu, ale fluktuace rychlosti v podélném směru jsou potlačeny pouze v jádru proudění, kde je také vyšší viskozita způsobená propojenou micelární strukturou. V oblasti u stěny jsou micely více orientovány ve směru proudění a podélné fluktuace rychlostí jsou stejné jako ve vodě. Je ukázán rozdíl mezi frekvencemi a amplitudami fluktuací ve vodě a ve vodě s aditivem. Rychlostní profil v trubici má ploché jádro s bodovými rychlostmi většími než ve vodě a oblast u stěny s rychlostmi mnohem menšími. Bezrozměrný rychlostní profil $u^+(y^+)$ se liší od profilu v polymerech, rovněž význačných ztráty zmenšujících aditivech. Bezrozměrný profil PAL závisí na Reynoldsově čísle a vyznačuje se větším sklonem než v polymeru a horizontálním zakončením.

Tlakové ztráty se obvykle interpretují jako závislost součinitele tření na Reynoldsově čísle, které je založeno na viskozitě vody. V grafech jsou tyto funkce silně závislé na průměru potrubí. Snížení tření je vymezeno do určité oblasti Reynoldsových čísel, existuje až do hodnoty Re, kdy smykové napětí překročí určitou mez a účinnost náhle končí. Po zmenšení smykového napětí se však schopnost snižovat tření opět obnoví. To je vidět na čárách $f(\mathrm{Re})$, které se se zvětšením průměru posunují do vyšších hodnot Reynoldsových čísel.

Tlakové poměry, vynesené v Prandtlových-Karmanových souřadnicích, se projevují jako kvasi-rovnoramenné trojúhelníky. Naše vlastní měření a měření některých jiných autorů ukazují, že při optimálních podmínkách může být snížení tření způsobené PAL větší než v polymerním roztoku. Obálka dosažených největších snížení tření je vyznačena jako přibližná asymptota maximálních snížení tření v roztocích PAL (mdras) pro různé průměry potrubí.

Koncentrace micelárního aditiva vyšší než je optimum pro daný průměr potrubí nemá vliv na další zmenšení tření, může pouze zvětšit rozsah působení do vyšších hodnot Re.



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*Seznam použitých symbolů*

$D$ – průměr potrubí [m],
$f$ – koeficient tření,
$l$ – délka potrubí [m],
$\Delta p$ – rozdíl tlaků [N],
$r$ – vzdálenost od osy [m],
$R$ – poloměr trubice [m],
$u$ – průměrná bodová rychlost [m s$^{-1}$],
$u^+$ – bezrozměrná rychlost,
$u'$ – odmocnina ze čtverce střední hodnoty fluktuace rychlosti [m s$^{-1}$],
$v$ – průřezová rychlost [m s$^{-1}$],
$v_*$ – třecí rychlost [m s$^{-1}$],
$y^+$ – bezrozměrná vzdálenost,
$\mu$ – dynamická viskozita [Pa s],
$\nu$ – kinematická viskozita [m$^2$ s$^{-1}$],
$\rho$ – hustota [kg m$^{-3}$],
$\tau_w$ – třecí napětí na stěně [Pa].

*Indexy*

$f$ – tangenciální složka,
$r$ – radiální složka,
$z$ – osová složka.

# Differences in the Flow Behaviors of Polymeric and Cationic Surfactant Drag-Reducing Additives

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# Differences in the Flow Behaviors of Polymeric and Cationic Surfactant Drag-Reducing Additives

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Research on cationic surfactant drag-reducing additives has grown in recent years because of their repairability after shear degradation, making them suitable for potential applications in recirculation flows such as district heating and district cooling systems. Substantial differences between their flow behaviors and those of high-polymer, drag-reducing additives have been found. These include the influence of preshearing, the effect of mechanical shear on degradation, the influence of tube diameter, maximum drag-reduction effectiveness, and the shape of their mean velocity profiles. Examples of these different flow behaviors are described. The differences suggest that the mechanisms for causing drag reduction may be different for the two types of additives.

## Introduction

Drag reduction by additives is a flow phenomenon in which small amounts of an additive in a fluid cause a reduction in the turbulent friction compared with that of the pure fluid at the same flow rate. The drag-reduced flow remains turbulent, however, with a modified turbulent structure.

A reduction in energy loss in turbulent pipe flow of wood pulp fiber suspensions in water was reported by Forrest and Grierson (1931) more than 65 years ago. This first report of drag reduction went largely unnoticed (Radin, 1974; Nadolink and Haigh, 1995). Mysels and his associates (Mysels, 1949; Agoston et al., 1954; Mysels, 1971) found that the pressure drop in pipe flow for gasoline thickened by aluminum disoaps was lower than that of pure gasoline at the same flow rate. Because of war-time secrecy requirements, their results were not released until 1949 when a patent was issued listing a 1945 application date. At the First International Rheological Congress in 1948, Toms (1949, 1977) reported drag-reduction results on dilute solutions of high-molecular-weight poly(methyl methacrylate) in monochlorobenzene. He observed that, at constant pressure gradient, the flow rate could be increased by the addition of the polymer. Drag reduction is sometimes called the "Toms Effect". The Mysels and Toms results were the first in which drag reduction was recognized.

Since the first reports of drag reduction, a large number of researchers have worked in this area. Nadolink and Haigh (1995) compiled a bibliography on drag reduction by polymers and other additives. There are over 4900 references dating from 1931 to 1994.

The potential use of drag-reducing additives in recirculating turbulent flow systems such as district heating or district cooling systems has generated interest in finding suitable additives for recirculating systems. Such applications would require additives which either do not degrade with mechanical shear or extensional flows or will repair after degradation. Since effective high-polymer, drag-reducing additives are sensitive to mechanical degradation, there has been a great deal of interest in aqueous surfactant drag-reducing systems in the last decade as these additives are "repairable" after mechanical degradation. These and other differences suggest that there may be differences in their drag-reduction mechanisms. The differences range from the form of their viscosity−shear rate curves, their response to shear-degrading conditions, tube diameter effects, limiting drag-reduction asymptotes, and limiting mean velocity profile asymptotes.

This paper will illustrate these differences from our own results and from data in the literature and will relate them to the differences in the nature of the two types of additives.

## Experimental Section

Rheological measurements were made with rotational rheometers (Rotovisco RV 20 and CV 20 of Haake) using procedures recommended by Haake which provide for necessary corrections. The steady shear-flow curves were measured with a coaxial cylinder fixture (Figure 1), and the first normal stress differences were measured with a cone−plate system (Figure 2).

Mean velocity profiles were measured with a laser Doppler anemometry (LDA) system which was assembled from standard Dantec components and a Carl Zeiss argon-ion laser. With the use of LDA, there is always an uncertainty in the location of the light beam's cross section, which is not a point but a rhombus-like plane with side lengths of the order of several tenths of millimeters but well below 1 mm. The distance of the rhombus center from the tube wall has an uncertainty of about 0.1 mm.

Surfactants used were Habon G (hexadecyldimethyl-(hydroxyethyl)ammonium 3-hydroxy-2-naphthoate supplied by Hoechst AG) and Ethoquad T/13-50 (tallow tris(hydroxyethyl)ammonium acetate supplied by Akzo-Nobel) with sodium salicylate counterion. At very low concentrations, the amount of drag reduction increases with concentration, but it levels off at some concentration which depends on the surfactant chemical composition (Chara et al., 1993). All drag-reduction data shown here are at or near this saturation level.

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# SIMILARITIES AND DIFFERENCES IN DRAG REDUCTION BEHAVIOR OF HIGH POLYMER AND SURFACTANT SOLUTIONS


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*Abstract* - The two most widely studied types of drag reducing additives are high polymers and surfactants. Their turbulent flow behaviors have many similarities but significant differences exist in their maximum drag reducing asymptotes, the limiting slopes of their mean velocity profiles for maximum drag reducing solutions as well as the magnitudes of peak values of $v'/u^*$ and the peak locations. Stress deficits due to small values of Reynolds stresses have been observed for both types of additives but zero Reynolds stress profiles have been reported only for surfactant solutions. These differences indicate that the mechanisms of drag reduction for the two types of additives are different. The fact that mechanical degradation of surfactant systems is reversible while for high polymers it is irreversible make the former more versatile for applications.


## I. INTRODUCTION

Drag reduction in turbulent flow was first recognized by Mysels, et al some fifty years ago [1,2,3]. They found the pressure drop in pipe flow for gasoline thickened by aluminum disoaps was less than that of gasoline at the same flow rate. Soon after, Toms [4,5] reported similar results with dilute solutions of high molecular weight polymethylmethacrylate in monochlorobenzene. Because of wartime security considerations, Mysels results were published after Toms'. In the past five decades turbulent drag reduction has been an active research field with over 4900 references [6], most of them dealing with high polymer drag reduction.

High polymers are very effective in reducing friction losses and have proven valuable in increasing flow rates in crude oil and other hydrocarbon pipelines most spectacularly in the 48-inch, 800-mile long Alyeska Pipeline from the North Slope in Alaska to Valdez. With currently available polymers, concentrations of 1 ppm in crude oil can give significant drag reduction [7].

Polymer additives are, however, susceptible to mechanical degradation and chemical bonds are broken irreversibly and the highest molecular weight, most effective molecules are the ones most sensitive to scission. Thus polymers lose their effectiveness when passing through a pump and additional polymer must be injected downstream of a pumping station to reduce friction losses in the next pipeline section. Polymers are therefore only useful in once-through applications. Fortunately polymer injection at only a few "bottleneck" sections of the Alyeska Pipeline was needed to increase throughput when North Slope production exceeded pipeline capacity [7].

Surfactant additives in water generally require higher concentrations than high polymers but their microstructures do reform quickly after mechanical degradation in pumps or in other regions of high shear. Surfactants have low molecular weights, of the order of hundreds, but can form long worm-like micelles which are believed to form 3-D network structures. While these structures are easily broken, they reform rapidly so that the surfactant solutions regain their effectiveness rapidly. Thus surfactants can be used in recirculation systems such as district heating or district cooling systems and research activity on surfactant drag reduction has grown appreciably in the past ten to fifteen years.

Catonic, nonionic, anionic and zwitterionic surfactants have all been shown to be effective drag reducing additives. Often a counterion is required to obtain good drag reduction.

This paper will discuss similarities and differences in drag reduction behavior between high polymer and surfactant systems. Significant differences in their behaviors indicate that the nature of their interactions with the turbulent flow field may be different. Since most studies have been done with cationic surfactants, this type of additive will be compared with high polymer additives.

## II. COMPARISONS OF HIGH POLYMER AND SURFACTANT DRAG REDUCERS AND OF THEIR TURBULENT BEHAVIORS

### A. Microstructure

The microstructures of polymers and surfactant drag reducing additives are quite different. Uncharged, flexible polymers form random coils in solution which may uncoil and elongate under shear or elongational forces. Polymers which contain charged groups are elongated even at rest. It is generally believed that surfactants that are effective drag reducers have worm-like or thread-like micelle structures. These systems may form three-dimensional networks which fully pervade the solution at rest or they may require shear to form networks.

### B. Onset of Drag Reduction

Figure 1, in which friction factor is plotted against generalized Reynolds number, illustrates the types of onset behavior observed in high polymer systems [8]. The solution is a 200 ppm polyethylene oxide (400,000 molecular weight) in benzene. Onset for polymer drag reduction occurs when a critical shear rate is reached. This shear rate decreases with molecular weight, concentration, goodness of the solvent and, for coiled polymers, the flexibility of the polymer solution chain [9]. If the critical shear rate occurs in the laminar flow region, no sharp onset is observed but only a gradual departure from the laminar line as Reynolds number increases (see Fig 1, 0.833mm ID tube). Liaw et al [9] called this "concentrated" polymer solution drag reducing behavior. For the larger tubes, onset is observed as departures from the Von Karman turbulent friction factor curve at higher Reynolds numbers when a critical shear rate is exceeded. Decrease of polymer concentration in a single tube yields similar changes in onset behavior, ie increase in the critical Reynolds number. "Concentrated" and "dilute" drag reducing behavior correspond to Type B and Type A behavior noted by Virk [10].

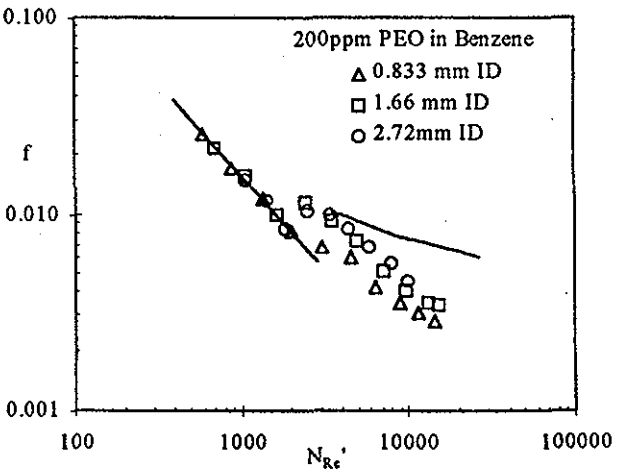


Figure 1. Effect of tube diameter on onset of polymer drag reduction

Almost all surfactant drag reduction friction factor data reported show gradual departure from the laminar friction factor line. There are a small number of reports of onset in the turbulent region. For example Gyr and Bewersdorff [11] show an example of a dilute surfactant solution (2mM of tetradecyl trimethyl ammonium bromide and sodium salicylate with 2mM sodium bromide added) giving onset in the turbulent regime.

C. Mechanical Degradation

As noted earlier, the high molecular weight polymers which are the most effective drag reducers, are also the most susceptible to chain scission in shear fields [12]. Extensional flows are even more effective than shear flows in causing degradation. Many investigators have reported on shear degradation of high polymers and some on the effects of predominantly extensional flows. Once a primary chemical bond has been broken, there is almost no chance that it will reform so polymer degradation is irreversible. Thus polymers can only be effective in once-through operations.

Surfactant micellar structures, on the other hand, rapidly self-assemble after mechanical degradation. The easily broken up microstructures reassemble in times of the order of seconds. Thus these additives have been successfully field tested in district heating and district cooling recirculation systems providing energy savings of 30 percent or more[13-16].

D. Maximum Drag Reduction Asymptotes

Virk demonstrated that for high polymers, a maximum drag reduction asymptote exists [17]. However, a number of experimenters, using surfactant and aluminum disoap drag reducing additives, have reported friction factor data lying significantly below Virk's limiting drag reducing asymptote. Zakin et al [18] examined these data as well as their own and proposed a new limiting asymptote for surfactant and aluminum disoap systems. The two asymptotes are shown in Figure 2. Virk's equation for the high polymer asymptote is [17]:

$$1/\sqrt{f} = 19.0 \log_{10}(N_{Re}\sqrt{f}) - 32.4 \qquad (1)$$

or

$$f = 0.58 N_{Re}^{-0.58} \quad (\text{for } N_{Re} = 4{,}000 \text{ to } 40{,}000) \qquad (2)$$

where f = friction factor and $N_{Re}$ = Reynolds number.

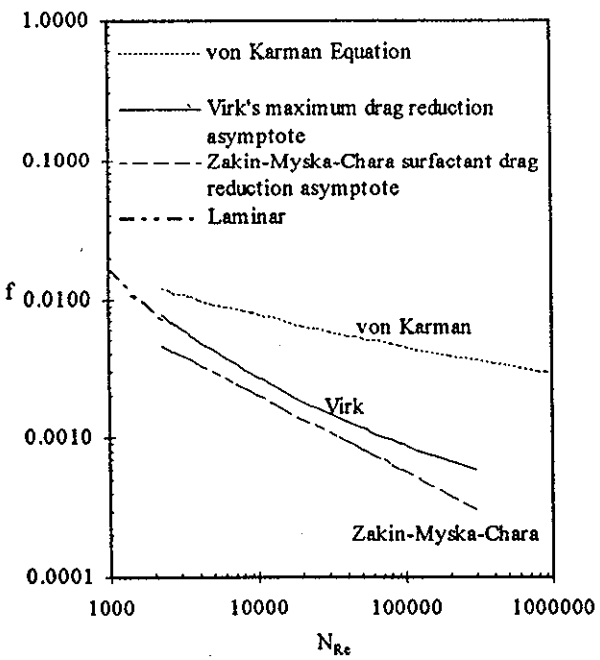


Figure 2. Friction factor vs Reynolds number

The limiting asymptote equation for surfactant and aluminum disoap solutions is [18]:

$$f = 0.32\, N_{Re}^{-0.55} \qquad (3)$$

This equation predicts friction factors more than 40% lower than Eq. 2 in some regions.

E. Mean Velocity Profile

For high polymer drag reducing systems, Virk proposed an elastic sublayer model with a limiting mean velocity profile equation:

$$u^+ = 26.9 \log_{10} y^+ - 17 \qquad (4)$$

This limiting velocity profile is consistent with the maximum drag reduction asymptote for high polymers (Eq. 1).

Recently, Zakin et al [18] offered another limiting profile equation reflecting the steeper slope they observed in the intermediate region for surfactant solutions approaching the maximum drag reducing asymptote for surfactants (Eq. 3) and also for aluminum disoaps in hydrocarbons:

$$u^+ = 53.9 \log_{10} y^+ - 65 \qquad (5)$$

where $u^+ = u/u^*$, u = local mean velocity, $u^*$ = friction velocity, $y^+ = yu^*/\nu$, y = distance from the wall and $\nu$ = kinematic viscosity. This limiting slope for surfactant and aluminium disoap solutions is twice that for high polymers. The lower limiting drag reducing friction factor asympotote for surfactants, Eq. 3, is a consequence of this steeper slope.

A schematic illustrating the two limiting equations is shown in Figure 3. Eq. 5 indicates a mixing length constant half of that of Eq. 4.

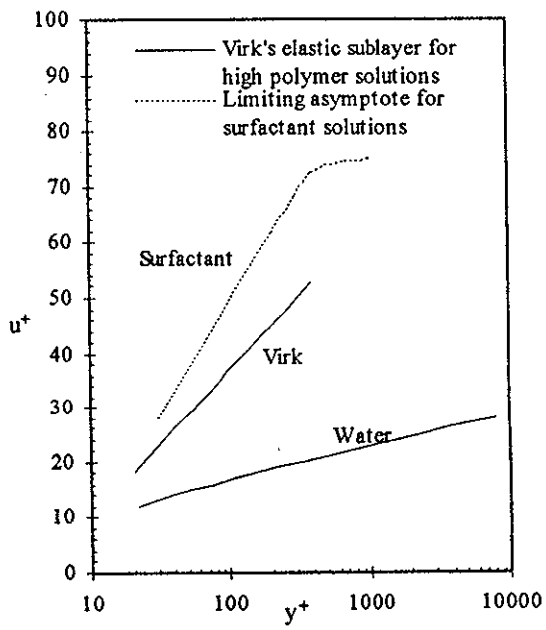


Figure 3. Mean velocity profiles ($u^+$ vs $y^+$)

F. Turbulence Measurements

1. *Axial Intensity Measurements*

A number of investigators have measured axial turbulence intensities of drag reducing fluids in channels and in pipes. Unfortunately, direct comparisons of the two types of additives in the same test systems are not available but some comparisons can be made.

Root mean square axial turbulence intensities at the center of the channel or the pipe for drag reducing polymer and surfactant solutions are about equal to or lower than for Newtonian solvents at the same Reynolds



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number [19]. Peak values of u'/u* near the wall of about 2 to 4 were observed for most drag reducing systems compared with 2.5 to 3.0 for the solvents. However, a peak value of 8 was reported by Rudd [20] for a polymer solution in a channel and by Bewersdorff and Ohlendorf [21] for a surfactant solution in a pipe at very high Reynolds number. Peak values increase with Reynolds numbers, as clearly shown in the data for a drag reducing surfactant solution of Schmidt [22], T.J. Hanratty's student. In contrast, his peak water intensities decreased slightly with Reynolds number.

In the data shown by Gampert and Rensch [19] and others, the u'/u* peak occurred at higher $y^+$ values for drag reducing systems than the $y^+ \approx$ 10-15 location typical of solvent data. Peak location is relatively insensitive to Reynolds number for solvents and for drag reducing systems.

2. *Transverse or Radial Intensity Measurements*

Radial intensities in the core region are lower for drag reducing solutions than for Newtonian solvents, both in absolute intensities, v', and in normalized intensities, v'/u* [22-24]. While maximum intensities occur at $y^+ \approx 100$ for Newtonian solvents, for drag reducing polymer solutions Gampert and Rensch [19] show a shift of the maximum intensity of v'/u* to somewhat higher values of $y^+$ with modest decrease in peak intensities. Schmidt [22], on the other hand, shows peak intensities reduced by 65% to 90% compared to water for a surfactant drag reducing system. Peak values for this system were shifted to lower $y^+$ values than those for water.

3. *Reynolds Stresses*

Wei and Willmarth [25] found 'large' negative Reynolds stresses in the near wall region of their channel into which concentrated PEO solutions were injected and small Reynolds stresses across the profile. Earlier, Durst et al [26] had also observed negative Reynolds stresses close to the wall. Schummer and Thielen [27], Willmarth et al [28] and Bewersdorff [29] had earlier noted stress deficit profiles based on their measurements of Reynolds stresses and mean velocity profiles. A stress deficit can also be seen near the wall in the earlier data of Patterson et al [30] for a high molecular weight polyisobutylene in mineral oil drag reducing solution.

Schmidt's [22] Reynolds stress measurements in a cationic surfactant drag reducing solution (Ethoquad T/13-50 — Sodium Salicylate) in a channel showed nearly zero Reynolds stress profiles at Reynolds numbers of 19,060, 29,750 and 49,130. This result, like those mentioned above, requires postulation of an additional (viscoelastic) stress term of significant magnitude. Kawaguchi et al [31, 32] obtained similar zero Reynolds stress profile results with a different cationic surfactant solution in a channel. The zero values are probably caused by a combination of low v' values of these surfactant solutions and phase differences between the u' and the v' intensities. Conditioned sampling of Reynolds stress data for these surfactant systems is needed to determine the magnitudes of the four possible combinations of u' and v' values. This would clarify how and why they sum to zero Reynolds stress and also the behaviors of turbulent sweeps and ejections in these systems.

## III. CONCLUSIONS

While there are similarities in the drag reducing behavior of high polymer and surfactant systems, there are significant differences, most notably the limiting asymptotes for maximum drag reduction and the ultimate slopes of the mean velocity profiles. Other differences are the larger reductions in peak v'/u* values for surfactant drag reducing solutions and a shift in the location of the peak value to lower $y^+$ compared to Newtonian solvents while the polymer solution peaks shifted to higher $y^+$.

Both types of additives have demonstrated a stress deficit, but near zero Reynold stress profiles have been observed in surfactant solutions, but not in polymer solutions. These differences indicate that there is some difference in the mechanism(s) for drag reduction for the two types of additives most probably because of differences in the interactions between turbulent eddies and elongated polymer molecules and their interactions with surfactant networks.

Finally, the ability of surfactant solutions to recover after mechanical degradation while high polymers are irreversibly degraded, allows the former to be used in a wider variety of applications including recirculation systems.

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ORIGINAL CONTRIBUTION

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# Micellar size of drag reducing cationic surfactants

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**Abstract** Dynamic light scattering was employed to determine the effects of surfactant nature, concentration and counterion ratios, and shear on the hydrodynamic radii of micelles of commercial cationic surfactants which are powerful drag reducers in turbulent flow at high temperatures. Such surfactants are potentially useful for reducing pumping energy losses in district heating and cooling systems.

**Key words** Surfactant micelle size – hydrodynamic radius – shear history – dynamic light scattering

## Introduction

Intensive research has been carried out on the influence of certain cationic, anionic and non-ionic surfactants, in some cases with counterion salts, on drag reduction in turbulent flow in pipes. The observed drag reduction has been ascribed to the presence of rod-like micelles in the solution. Surfactant drag reducers may lose their drag reduction ability when subject to high shear but quickly regain their effectiveness when flowing in a regime of lower shear, allowing them to be used in recirculation systems where polymer additives which mechanically degrade irreversibly cannot be used. This is believed to be due to the ability of micelles to form long threads or networks which break up under high shear but reform when shear is removed.

During the last few years much effort has been given to developing cationic surfactants which are effective over a wide range of temperatures such as are encountered in district heating or cooling systems. Quite a few results have already been published on physical–chemical, hydraulic and rheological properties of such surfactants and their possible usage, see for example refs. [1–5]. In this paper, we want to show and discuss one of the principal properties of single micelles, namely their size.

Ohlendorf et al. [6] showed that the length of rod-like micelles of hexadecyl trimethyl ammonium bromide with sodium salicylate ($C_{16}$ TA Sal) determined by electric birefringence decreases with rising temperature and increases with surfactant concentration. Micelle lengths increased from 30 nm to 70 nm at 30 °C when concentration increased from 0.2 to 0.4%. At 50 °C, the lengths were 18 and 30 nm at the same concentrations. For tetradecyl trimethyl ammonium salicylate ($C_{14}$ TA Sal) micelles at 30 °C, the lengths were 30 and 40 nm at the same two concentrations. Kalus et al. [7] reported that micelles of 5 mM tetradecyl trimethyl ammonium salicylate in $D_2O$ at 25 °C are approximately 30 nm long and their diameter is approximately 4 nm based on small angle neutron scattering measurements. Using electrical birefringence measurements, Bewersdorff and Ohlendorf [8] found that the length of micelles of $C_{14}$ TA Sal decreases with decrease in salt content and with increase in temperature.

Interesting results on cetyl trimethyl ammonium/sodium salicylate micelles were reported by Nemoto and

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With the exception of Habon G, the distributions of relaxation times (and therefore also the distributions of other quantities) for fresh samples were observed to be unimodal which means that the solutions contained only one kind of particle, but the distributions were not monodisperse, the width depending on the concentration.

## Results and discussion

### Concentration influence

The dependence of *Rh* of micelles in ET13/NA in distilled water on the surfactant concentration (at fixed molar ratio) of both components and the distribution of *Rh* at a concentration 1.25/1.87 mM are shown in Figs. 2 and 3. (This molar concentration corresponds to a total concentration of ET13 of 1.135 g/l (50% active matter) and 0.3 g/l of NA). *Rh* increased from 20.5 nm to 25.5 nm when concentration increased from 1.25/1.87 mM to 5/7.5 mM. This 25% change is not large for the fourfold concentration range. Extrapolation to infinite dilution yields $Rh = 19.5$, but is not physically meaningful as rod-like micelles would not form at low concentration. The distributions of hydrodynamic radii are rather large. At 1.25/1.87 mM, *Rh* ranges from 11 nm to 46 nm and the width, *w*, of the distribution is 61% of a decade. The estimated width increases with concentration, showing a 45% increase at 5.0/7.5 mM as listed in Table 1.

Measurements of 3 mM ET13 with 5.0, 7.5 and 12.5 mM NA in distilled water gave an increase of the mean hydrodynamic radius of only 10% (Figs. 4 and 5). The distilled water samples contained some impurities and for that reason each was measured twenty times. Only results which were not affected by scattering from impurities are shown. The width of the distributions decreased by 33% as NA/surfactant ratio increased. Thus, with decreasing ET13/NA ratio, the dispersion of the micelles tends to monodispersity.

Similar influence of the salt content in the sample was found with AR/NA solutions in distilled water and shown in Figs. 6 and 7. Table 2 shows the increase of mean *Rh* values and the decrease of *w* values with the decrease of surfactant/salt ratio, i.e., with increase of sodium salicylate content, C(NA), at constant concentration of the surfactant, C(AR):

In this case there is a 13% increase of *Rh* over a 1.9 fold concentration ratio while *w* decreases 29%. The extrapolation to zero concentration C(NA) yields $Rh = 27$ nm.

The results with AR/NA and ET13/NA solutions are very similar though their molecular weights differ considerably. In general, these results showing the influence of the concentration of the mixture and that of the ratio of surfactant/salt are in accordance with results from the investigations using pure chemicals presented in refs. [6–8, 10–13].

Another effective drag reducing surfactant is ETO with a molecular weight between those of AR and ET13. ETO/NA solution with 3.0/12.5 mM concentration in distilled water has an average mean hydraulic radius of

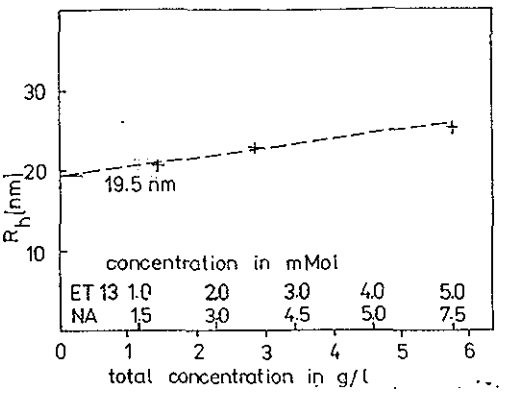


Fig. 2 Dependence of hydrodynamic radius, *Rh*, of the solution of Ethoquad T13/50 with sodium salicylate on total concentration of both components; the ratio of concentrations of both components C(ET13)/C(NA) is constant

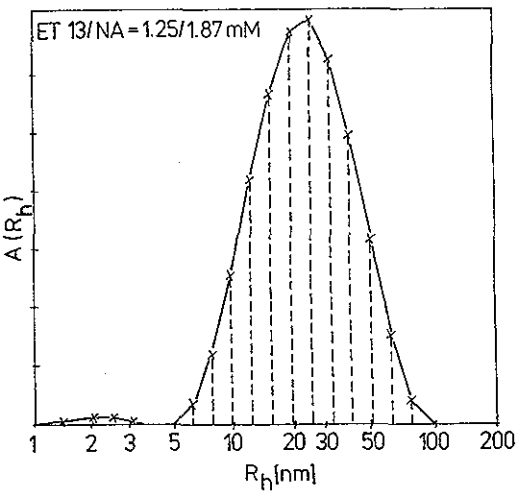


Fig. 3 Plot of scattered light, *A*(*Rh*), vs. hydrodynamic radius, *Rh*, in ET13/NA (1.25/1.87 mM) solution in distilled water

**Table 1** Effect of ET13/NA Concentration on *Rh* and *w*

| Surfactant/NaSal Concentration | *Rh*, nm | Distribution Width, *w* In% |
|---|---|---|
| 1.25/1.87 mM | 20.5 | 61% |
| 2.50/3.75 mM | 23.0 | 70% |
| 5.00/7.50 mM | 25.5 | 88% |

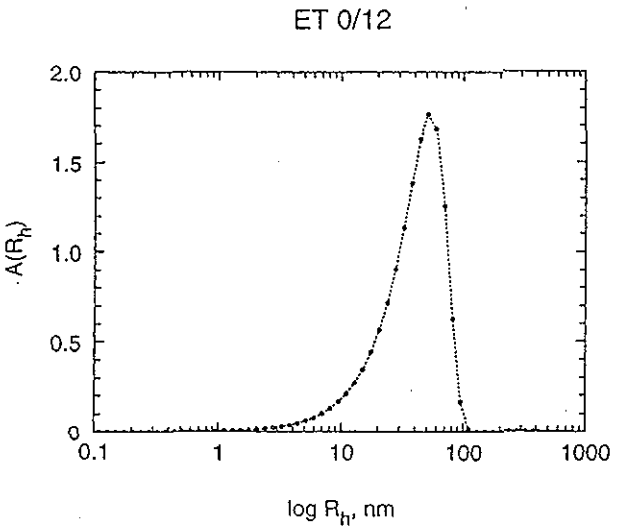


**Fig. 8** Distribution function of Ethoquad 0/12 with sodium salicylate in distilled water; the concentration of ET0/NA = 3/12.5 mM

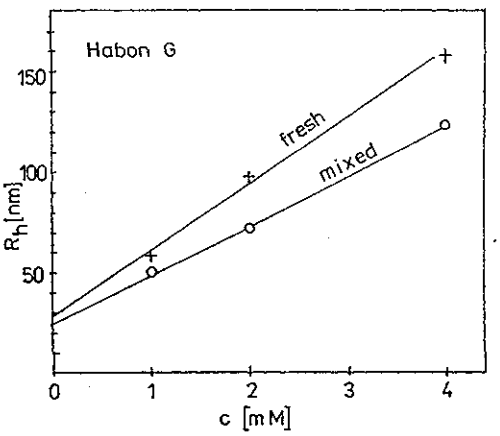


**Fig. 9** Dependence of *Rh* on concentration of Habon G

influence of intense shearing by mixing in a beaker for two hours (which was applied to simulate the effect of pumping on the breakdown or degradation of the micelles) were investigated. Light scattering measurements were made several days after mixing.

Hydraulic radii as a function of concentration for the fresh and the sheared solutions are plotted in Fig. 9. Over a fourfold range of concentration (1 to 4 mM), *Rh* shows a strong dependence on concentration, varying from 58 nm to 157 nm in the fresh solution which is a 170% increase. The growth of micelle size with concentration is large compared with other surfactants. Intensive shear reduced the hydraulic radii of the micelles by 16 to 28%, the effect of shear being more pronounced with the larger, higher concentration micelles. If we calculate a volume

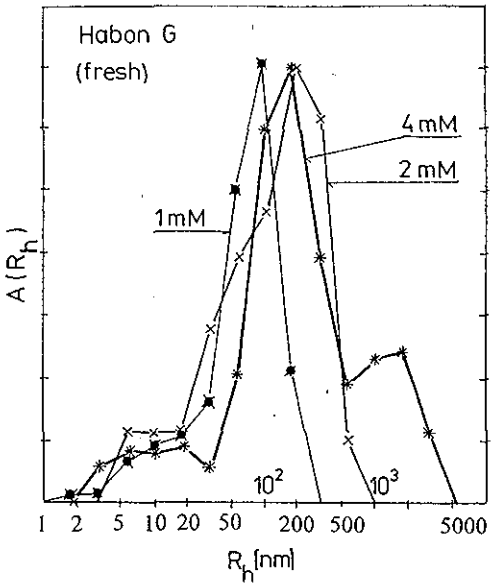


**Fig. 10** Distribution function in fresh Habon G solutions of 1, 2 and 4 mM concentrations in distilled water

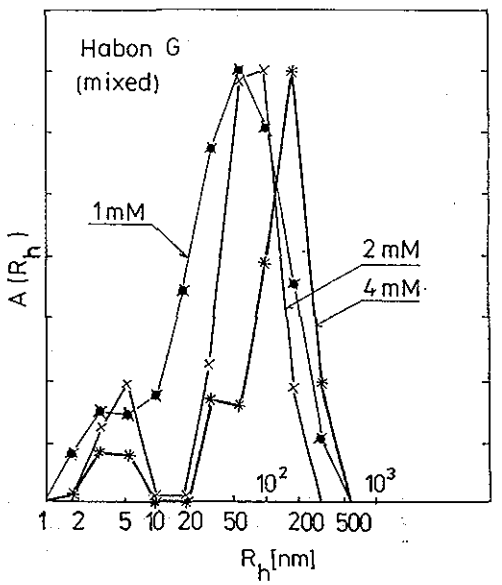


**Fig. 11** Distribution function in sheared Habon G solution of 1, 2 and 4 mM concentrations in distilled water

change of hypothetical spheres with diameter 2*Rh*, this decrease of *Rh* corresponds to a significant 41 to 63% decrease in volume of the micelles. In the limit of infinite dilution both series of samples (fresh and mixed solution) approach a hydrodynamic radius, *Rh*~25 nm, which is close to the value obtained with ET13/NA.

The particle size distributions for the fresh and degraded Habon G solutions are shown in Figs. 10 and 11.

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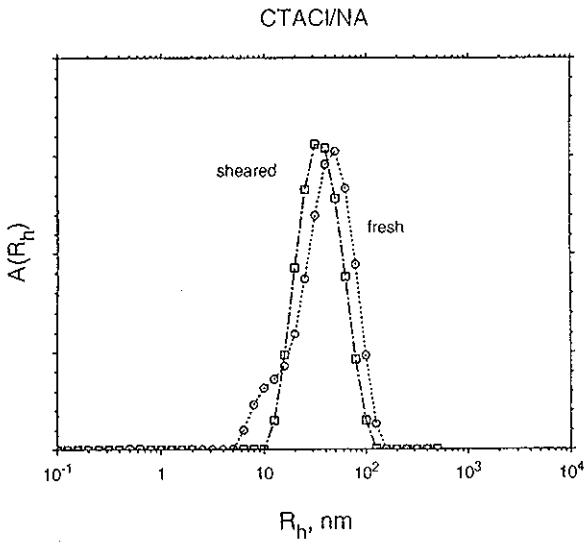


**Fig. 14** Distribution functions in CTAC/NA solutions with 1.25/12.5 mM concentration in distilled water, the influence of shearing

**Table 3** Effect of Shear on *Rh* and *w*

| | *Rh*, nm | Distribution width, *w*, In% |
|---|---|---|
| ET13/NA = 5/12.5 mM | | |
| Fresh solution | 35.4 | 107% |
| Sheared solution | 35.8 | 101% |
| AR/NA = 5/12.5 mM | | |
| Fresh solution | 33.5 | 107% |
| Sheared solution | 31.6 | 80% |
| CTAC/NA = 1.25/12.5 mM | | |
| Fresh solution | 31.6 | 58% |
| Sheared solution | 29.3 | 57% |

with average lengths of 90–130 nm. Polydispersity can also be seen in the micrographs.

The hydrodynamic radii of ET13/NA micelles obtained from these light scattering measurements range from about 20–35 nm. These radii are consistent with the electron microscope lengths given in [20] as Hu, Wang and Jamieson [14] estimate that the relation between *Rh* and the length of the micelle gives a micelle length approximately four times *Rh*.

## Conclusions

1) Micelle size, represented by hydrodynamic radius, increases with the concentration of the solution as well as with increase in counterion concentration. *Rh* increases when tap water replaces distilled water.

2) Micelles of Habon G are approximately four times larger than micelles of Ethoquad T13 or Arquad 16. Habon G solutions reach maximum drag reduction effectiveness at about one-fifth of the concentration for the other surfactants.

3) Considerable polydispersity was found in these surfactants. A purer sample similar in composition to AR had similar average size but a narrower distribution than the commercial sample.

4) Intensive shearing (mixing) reduces the size of large micelles in Habon G solutions while the sizes of the smaller micelles in solutions of Ethoquad T13, Arquad 16 and pure CTAC are not greatly affected by shearing.

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