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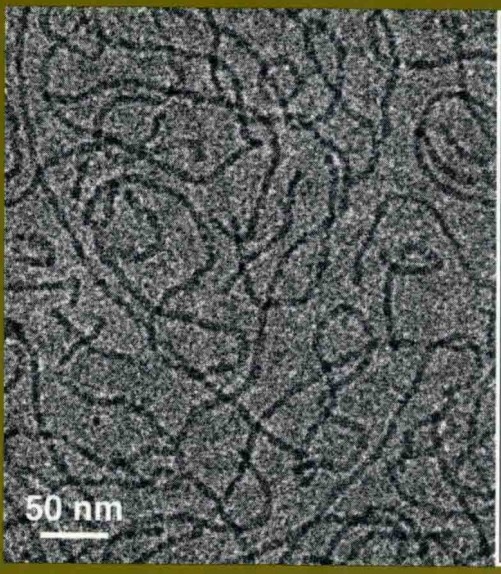
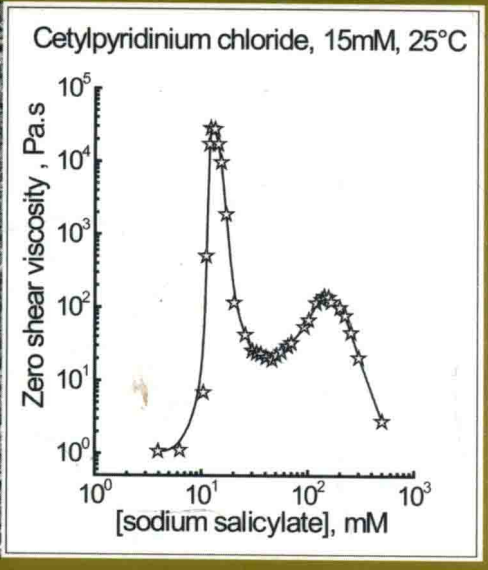
GIANT MICELLES

Properties and Applications





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GIANT MICELLES

PROPERTIES AND APPLICATIONS

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Introduction

Amphiphilic molecules such as surfactants have the remarkable ability to self-assemble in aqueous solutions to form a variety of microstructures including liquid crystals, vesicles, small micelles, and of particular interest here, long worm- or threadlike "giant" micelles. The history of the physical chemistry of micellar solutions dates back to a paper by J.W. McBain[1] in 1913, in which he proposed the then-controversial idea that self-assembled aggregates could form in aqueous solution. New ideas in science are often greeted skeptically, and this was no different. When, at a meeting of the Royal Society of Chemistry, McBain pointed out that changes in the colligative properties of surfactant solutions around the critical micelle concentration could be explained by the formation of micelles, the distinguished meeting chair responded,[2] "Nonsense, McBain." Scientists persevered, nonetheless, and micellization is now a widely studied phenomenon.

The giant micelles discussed in this volume refer to the long micelles that occur in many types of surfactant solutions, most frequently upon increasing the concentration of the surfactant or of an added salt, or in the presence of specific counterions. These micelles may be microns long and have either a circular or elliptical cross section of a few nanometers. These micelles can be discrete "worms" or "threads," or may crosslink into a network. In all cases the micelles can break and reform spontaneously or under stress, and so are often called "living polymers." To the best of our knowledge the expression "giant micelles" was first coined by G. Porte[3] in 1983.

The transition from spheroidal micelles to giant micelles has been directly visualized by transmission electron microscopy at cryogenic temperature, and the images show coexistence of spherical and elongated micelles. Giant micelles also form in organic liquids and on solid surfaces. The rheological behavior of solutions containing giant micelles has been studied in detail, and these rheological properties are the key to the many successful applications of these solutions. Giant micelles are *equilibrium* or *living polymers* as the micelle molecular weight fluctuates with time, and so are distinct from conventional polymers where the repeat units are covalently bonded and the molecular weight is constant. In some cases the giant micelles can close upon themselves (forming *ringlike* micelles) or *branch* rather than continue to grow in one dimension when a parameter (e.g., temperature or salt concentration) is modified. These branched micelles can ultimately form a network of micellar junctions. All these possible behaviors give rise to a variety of phenomena that have been and still are the subject of extensive investigations. Recent reports show that giant micelles can be formed by amphiphilic block copolymers, or by surfactants synthesized to respond to external stimuli by reversibly forming giant micelles from spheroidal micelles.

If the expression "giant micelle" is relatively recent, the concept of very long and linear micelles is surely not. The first experimental study, by light scattering, of elongated or "rodlike" micelles was reported by P. Debye and E.W. Anacker[4] in 1951. They concluded that the variation of the light measured as a function of scattering angle (the dissymmetry) agreed only with the presence of long rodlike micelles. They said:

> The micelles of n-hexadecyltrimethylammonium bromide in ... potassium bromide are large enough to produce measurable dissymmetry in the scattered light. Dissymmetry measurements showed conclusively that these micelles are not spherical or disk-like in shape; analysis of the data indicates that the micelles are rod-like. The cross section of such a rod will be circular, with the polar heads of the detergent lying on the periphery and the hydrocarbon tails filling the interior. The ends of such a rod would most certainly have to be rounded off with polar heads.

Thus, both the concept of one-dimensional growth and the importance of micellar "endcaps" were established.

The impact of that micellar microstructure on flow or rheological properties of surfactant solutions was also noted early on, and was discussed in 1954 during a report of the study of the viscoelasticity of surfactant solutions. The following excerpt from a paper by N. Pilpel[5] in 1956 compels some modesty about recent achievements in the study of giant micelles. He said:

> At low concentration of electrolyte the soap (sodium oleate) is believed to be in the form of small spherical micelles but at higher electrolyte concentration these pack together into cylinders which then interlink to form a network structure.

This single short sentence describes both the sphere-to-rod transition of micelle shape and the formation of networks in solutions of giant micelles. Both topics have been investigated at great length and described in probably thousands of papers since. Two names emerge from the large number of workers involved in this effort: Candau[6] was the first to point out the analogy in the rheological behaviors of giant micelles and polymers; Hoffmann[7] was the first to emphasize that this rheological behavior can be completely dominated by the rate of reversible scission of giant micelles.

One of the important reasons that this field of study has been able to move forward quickly is that a powerful theoretical framework has evolved in parallel with a range of sophisticated experimental tools. The theoretical approaches also reach back to P. Debye,[8] who suggested that the thermodynamics of the micelle would reflect a balance of repulsions between the (ionic) surfactant head groups and van der Waals's attractive forces between the hydrocarbon tails. Although couched in terms of an incorrect lamellar micellar geometry, this "opposing force model" provided a seed from which much more detailed thermodynamic models have grown over the past 50 years. Of particular importance is the work of C. Tanford and coworkers[9] in the 1970s, who introduced a robust thermodynamic framework and used it to make a strong connection between micelle formation and the more general hydrophobic effect.

Theory has also contributed significantly to our understanding of the rheological properties within the framework of a mean field model developed by M. Cates[10] and co-workers. They opened the way to interpreting measurements of rheological properties by showing how the dynamic processes of micellar diffusion and the kinetics of micellar breakage and recombination can interact to set the time scales for stress relaxation in entangled wormlike micellar solutions. In many cases there is only one characteristic relaxation time, so many of the rheological properties can be interpreted in terms of the remarkably simple Maxwell model of a viscoelastic fluid.

The variety of behaviors encountered in solutions of giant micelles, the present and past interest in these systems, and their applications in important industrial processes led us to the idea of summing up in a volume the "state of the art" in this area of science. The present volume comprises three parts. The first four chapters focus on theoretical aspects of the formation of giant micelles from different points of view (molecular thermodynamics, packing, and computer simulations) and of their rheological behavior. The following 10 chapters deal with experimental aspects: transmission electron microscopy at cryogenic temperature, scattering methods, phase diagrams, linear and nonlinear rheology, and relaxation. They also review the properties of giant micelles on solid surfaces and systems of smart micelles that respond to external stimuli by a change of shape. Another chapter describes giant micelles formed from amphiphilic block copolymers. The last chapter in this part reviews noncovalent polymers stabilized by hydrogen bonds that show rheological behavior very similar to that of giant surfactant micelles. The third part comprises four chapters that review the applications of giant micelles in oil and gas production (a growing application), in drag reduction (an energy saving process), in shampoos, and in other products used by consumers daily, such as hard surface cleaners, personal care products, sunscreens, and drug delivery formulations.

We believe that having this range of information together in one volume will be of considerable help to the community studying and using these fascinating materials.

The Editors

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Editors

Dr. Raoul Zana is directeur de recherches emeritus at the Institut C. Sadron of the CNRS. He earned his degree in chemistry from the University of Strasbourg and that of *ingenieur chimiste* from the Ecole Nationale Superieure de Chimie de Strasbourg, both in 1958. He obtained his Doctorate es Sciences in 1964 at the Centre de Recherches sur les Macromolecules (CRM) of the CNRS, working with Professors R. Cerf and H. Benoit on ultrasonic relaxation in polymer solutions. From early 1965 to mid-1967 he was a postdoctorate investigator at Case Western Reserve University (Cleveland, Ohio), working with Professor E. Yeager. There he brought to use a method of measurement of ultrasonic vibration potentials, an effect predicted by P. Debye. This work was in part at the origin of the now widely used method of measurement for zeta potentials of colloidal systems. He returned to Strasbourg at the CRM (then Institut Charles Sadron) as a *director de recherches* and started his work on polyelectrolytes and micellar systems. He created the research group "physicochemistry of colloids" and directed it for many years.

Dr. Zana has directed for 4 years the so-called Coordinated Research Group "Microemulsions" that included 80 scientists. He has served on the editorial boards of the journals *Langmuir*, *Journal of Colloid and Interface Science*, *Current Opinions in Colloid Interface Science*, and *Southern Brazilian Journal of Chemistry*. He has held many appointments as an invited professor: University of Sherbrooke (Canada), Drexel University (Philadelphia, Pennsylvania), Science University (Tokyo), University of Palermo (Italy), Technion (Haifa, Israel), University of Fukuoka (Japan), University of Auckland (New Zealand), University of Sydney (Australia), and California State University (Northridge, California). He has been consulting with several companies and has served in various positions in the CNRS and the University L. Pasteur of Strasbourg. He has published more than 375 papers, reviews, or chapters in volumes, and edited three volumes of the Surfactant Science Series: *Surfactant Solutions: New Methods of Investigation*, *Gemini Surfactants*, and *Dynamics of Surfactant Self-Assemblies*. He contributed much to each of these volumes. The last two volumes describe a good part of his studies.

Eric W. Kaler earned a BS degree in chemical engineering (with honors) from the California Institute of Technology in 1978 and a PhD in chemical engineering from the University of Minnesota in 1982 working with L. E. Scriven and H. T. Davis. Dr. Kaler joined the faculty of the Department of Chemical Engineering at the University of Washington in 1982 and was promoted to associate professor in 1987. He moved to the Department of Chemical Engineering at the University of Delaware in 1989, became a professor there in 1991, department chairman in

1996, and was appointed the Elizabeth Inez Kelley Professor of Chemical Engineering in 1998. He became dean of the College of Engineering in 2000. He was also a visiting professor at the Universität of Graz in Austria in 1995. His research interests are in the areas of surfactant and colloid science, statistical mechanics, and thermodynamics.

Dr. Kaler received one of the first Presidential Young Investigator Awards from the National Science Foundation in 1984, the Curtis W. McGraw Research Award from the American Society of Engineering Education in 1995, the 1998 American Chemical Society Award in Colloid or Surface Chemistry, and the 1998 ACS Delaware Section Award. He was elected a fellow of the American Association for the Advancement of Science in 2001, and received the Chilton Award from the Wilmington AIChE Section in 2002. In 2005, Dr. Kaler was awarded the E. Arthur Trabant Institutional Award for Women's Equity by the University of Delaware and received the Lectureship Award from the Division of Colloid and Surface Chemistry of the Chemical Society of Japan. He received the Kash Mittal Award from the Surfactants in Solution Symposium in 2006. He has chaired three Gordon Research Conferences and serves or has served on the editorial boards of the journals *Langmuir*, *Colloids and Surfaces*, *Colloid and Interface Science*, and *AIChE*, and was an associate editor of the *European Physical Journal*. He is also the founding coeditor-in-chief of the international journal *Current Opinions in Colloid and Interface Science*. He has authored or coauthored more than 200 peer-reviewed papers and holds 10 U.S. patents. He has been a consultant to numerous companies, and has served in a variety of positions in several professional societies.

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