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Editorial

In recognition of the many contributions of James E. Mark to polymer science and technology

We are honoured to dedicate this special issue of Polymer to Professor James E. Mark in recognition of his many outstanding contributions to polymer science and technology and on the occasion of his 70th birthday. His contributions span more than 40 years and are documented in nearly 650 publications beginning with his first paper with Paul Flory in 1963.[1] His pioneering work in the theory of rubber elasticity and silicone chemistry has been recognized by many major national awards from the American Chemical Society including the George Stafford Whitby Award in 1991 and the Charles Goodyear Medal in 1999 from the Rubber Division of the American Chemical Society, the ACS Applied Polymer Science Award in 1994, the Flory Polymer Education Award (ACS Division of Polymer Chemistry) in 2000, and the Kipping Award in Silicon Chemistry in 2004. Professor Mark has also received the Award for Outstanding Achievement in Polymer Science and Technology from the Society of Polymer Science, Japan, in 2004.

James E. Mark was born on December 14, 1934 in Wilkes-Barre, Pennsylvania. He received his BS degree in 1957 in Chemistry from Wilkes College and his PhD degree in 1962 in Physical Chemistry from the University of Pennsylvania. After serving as a Postdoctoral Fellow at Stanford University under Professor Paul J. Flory, he was Assistant Professor of Chemistry at the Polytechnic Institute of Brooklyn before moving to the University of Michigan, where he became a Full Professor in 1972. In 1977, he assumed the position of Professor of Chemistry at the University of Cincinnati, and served as Chairman of the Physical Chemistry Division and Director of the Polymer Research Center. In 1987, he was named the first Distinguished Research Professor, a position he holds at

[1] A complete listing of publications follows this preface.

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the present time. In addition, he has extensive research and consulting experience in industry and has served as a Visiting Professor at several institutions. Professor Mark's research interests pertain to the physical chemistry of polymers, including the elasticity of polymer networks, hybrid organic–inorganic composites, liquid–crystalline polymers, and a variety of computer simulations. Professor Mark has lectured extensively on polymer chemistry, has been an organizer and participant in a number of short courses, and has published approximately 650 research papers and co-authored or co-edited 20 books. He is the founding editor of the journal Computational and Theoretical Polymer Science, established in 1990, is an editor for the journal Polymer, and serves on a number of journal Editorial Boards. He is a Fellow of the New York Academy of Sciences, the American Physical Society, and the American Association for the Advancement of Science. He was elected to the Inaugural Group of Fellows (ACS Division of Polymeric Materials Science and Engineering). In addition to the major awards from ACS cited earlier, Professor Mark received the Dean's Award for Distinguished Scholarship, the Rieveschl Research Award, and the Jaffe Chemistry Faculty Excellence Award from the University of Cincinnati. Other important recognitions include the Turner Alfrey Visiting Professorship, the Reed Lectureship at Rensselaer, and the Edward W. Morley Award from the ACS Cleveland Section.

Professor Mark has made seminal contributions to many areas of polymer science and technology. These are reflected in the main sections of this issue that focus on rubber elasticity, molecular modeling, and in situ composites. His individual contributions to each of these areas are discussed below.

1. Contributions to the theory of rubber elasticity

Professor Mark has contributed enormously to our understanding of rubber elasticity throughout his distinguished career. Between the time of his PhD research with R.E. Hughes, his Postdoctoral studies with P.J. Flory, and the present, he has gifted us with a tremendous body of work. In the limited space available here, we would like to highlight just a few of the advances and contributions that Professor Mark has made.

The two books with Burak Erman, Rubberlike Elasticity. A Molecular Primer and Structures and Properties of Rubber like Networks, have provided both the novice and expert with much needed updates on the technology and theory of elastomers. The later book provides the most comprehensive survey of the current state of rubber elasticity research since the classical and defining work of Treloar.

Professor Mark's exploitation of bimodal networks to improve the modulus and toughness of polymers was a technological breakthrough. Especially useful for the relatively weak silicones, the method of incorporating short chains along with long ones into the network creates much improved properties. Similarly, his advocacy of hybrid materials, including those formed by in situ precipitation of fillers, has contributed tremendously in the development of an important array of new materials, as is seen in some of the contributions in this volume.

Professor Mark has done remarkable experimental research to elucidate the elastic equation of state. He has shown how finite chain extensibility and crystallization are both implicated in departures from Mooney–Rivlin behavior at large uniaxial extensions. Some of his most critically important work on the experimental elasticity was on the behavior of the stress–strain curve in the vicinity of the unstressed state. The difficult experiments in this region, where the experimental elastic system goes from uniaxial extension to biaxial extension, have misled more that a few experimentalists. The papers by Xu and Mark are critical reading for anyone interested in the details of this important transition region and the excursion into uniaxial compression.

2. Molecular modeling and molecular simulations

Professor Mark began his work with statistical mechanics of polymer and biopolymer chains in the early to mid-sixties at the start of his career during his postdoc with Paul Flory and later association with Murray Goodman at Brooklyn Poly and Sam Krimm at Michigan and has continued to the present time. Early work focused on the configurational statistics of all the major classes of flexible chain polymers including polysiloxanes, polyoxymethylene, polyoxyethylene, polytrimethylene oxide, polybutadiene, polyisoprene, polyethylene, ethylene–propylene copolymers, polyphosphazenes, polysulfides, and a variety of vinyl polymers. Beginning in 1981 at the University of Cincinnati, Professor Mark in collaboration with Bill Welsh extended their investigation to several important rod-like polymers including polybenzobisoxazole and polybenzothiazole. This large body of work provided the foundations for our current understanding of configurational properties.

Professor Mark's entry into the area of molecular simulation may be traced to several publications in 1990 that dealt with the use of Monte Carlo methods to study free volume in collaboration with Frisch, Roe, Rigby, DeBolt, Trohalaki, and Takeuchi, and the use of molecular dynamics in the simulation of diffusion of small molecules in collaboration with Trohalaki, Rigby, Kloczkowski, and Roe. In 1990 alone, Mark published 27 papers.

In 1991, Mark and Fried collaborated to publish a new journal focusing on the molecular simulations of polymeric materials (Computational Polymer Science). This journal later extended its scope to include theory and became Computational and Theoretical Polymer Science in 1996 and was later incorporated into Polymer in 2002.

3. In situ composites

In 1982, Mark and Pan reported that silicone elastomers could be reinforced with silica generated in situ via a sol–gel reaction. This discovery led to a rich new field that has attracted the attention of scores of researchers. Professor Mark immediately realized that at high silica loadings, reinforced silica was a polymer-toughened ceramic. In fact, a spectrum of mechanical properties could be achieved using chemistry to control the oxide/elastomer ratio. At that time, the nanomaterials vocabulary had not been invented, but there is no question that Professor Mark's contributions in reinforced elastomers inspired many researchers currently pursuing nanocomposites.

In addition to silica, Professor Mark's group investigated a host of oxides, zeolites, polyhedral oligomeric silsesquioxanes (POSS), and glassy polymers as novel reinforcing agents. Professor Mark came up with the idea of assigning 'tasks' to different constituents in order to overcome the usual trade-off between elongation and strength and to impart new properties to elastomers. Magnetic particles, for example, could be aligned and manipulated using magnetic fields.

In addition to developing a new class of materials, Professor Mark also led the effort to understand the properties of hybrid elastomers. This endeavour involved thorough characterization of the structure using scattering and electron microscopy supported by theoretical studies and simulations.

Professor Mark's prolific scientific contributions have come not only from the combination of an imaginative mind and boundless energy but also from effective collaborations. Hundreds of students, postdocs, colleagues and friends, including the editors of and contributors to this issue, are the beneficiaries of his open communications and cooperative character. It is an honour for us to recognize the contributions of Professor Mark to polymer science and thank him for the influence he has had on our careers.

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