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Gelation and Phase Separation in Aqueous Methylcellulose Solutions

ABSTRACT

It is well established that aqueous solutions of methylcellulose (MC) can form hydrogels on heating, with the rheological gel point closely correlated to the appearance of optical turbidity. However, the detailed gelation mechanism, and the resulting gel structure remain poorly understood. We recently discovered that MC chains associate into fibrils on heating. The fibrillar structure was precisely quantified with a powerful combination of (real-space) cryogenic transmission electron microscopy (cryo-TEM) and (reciprocal space) small-angle neutron scattering (SANS) techniques. The cryo-TEM images reveal that MC chains with a molecular weight of 300,000 g/mol associate into fibrils with a remarkably uniform diameter of 15 ± 2 nm over a range of concentrations. Vitrified gels also exhibit heterogeneity in the fibril density on the length scale of hundreds nanometers, consistent with the observed optical turbidity. The fibrillar nature of the gel structure was confirmed by fitting the SANS data consistently with a model based on the form factor for flexible cylinders with a polydisperse radius. This combination of complementary experimental techniques provides a comprehensive nanoscale depiction of fibrillar morphology for MC gels, which correlates very well with macro-scale rheological behavior and optical turbidity previously observed for such systems.

BIOGRAPHY

Tim Lodge was born in Manchester, UK, in 1954, and emigrated to the US in 1968. After graduating from Harvard in 1975 with a B.A. cum laude in Applied Mathematics, he completed his PhD in Chemistry at the University of Wisconsin with Professor John Schrag in 1980. Tim then spent 20 months as a National Research Council Postdoctoral Fellow at NIST, collaborating with Dr. Charles Han. Since 1982 he has been on the Chemistry faculty at Minnesota, and in 1995 he also became a Professor of Chemical Engineering & Materials Science. He was named a McKnight Distinguished University Professor in 2001, an Institute of Technology Distinguished Professor in 2004, and he held the Lloyd H. Reyerson Chair in Chemistry from 2007-2011. In 2013 he was named a Regents Professor, the University's highest academic rank.

He was co-recipient of the 1993 George Taylor Alumni Award for excellence in research, given by the Institute of Technology, and in 1994 he was named a Fellow of the American Physical Society (APS). He received the Arthur K. Doolittle Award from the Polymeric Materials Science & Engineering Division of the American Chemical Society (ACS) in 1998. He was a co-recipient of the Society of Rheology Publication Award in 2003, and in 2004 he received the APS Polymer Physics Prize, and the Paul Flory Research Award from POLYCHAR. In 2007 he was recognized with the Nelson Taylor Award in Materials Science from Penn State. He was elected to Fellowship in the American Association for the Advancement of Science in 2009, and he received the International Scientist Award from the Society of Polymer Science, Japan. He was the recipient of the 2010 Prize in Polymer Chemistry from the ACS, and was also elected a Fellow by the ACS in 2010. In 2012 he received the Minnesota Award from the Minnesota Section of the ACS, and the Postbaccalaureate, Graduate and Professional Education Award from the University of Minnesota.

From 1994-2000 Tim served as Regional Editor for *Macromolecular Chemistry and Physics*, and since 2001 he has been the Editor of the ACS journal *Macromolecules*. In 2011 he became the founding Editor for a new journal, *ACS Macro Letters*. He is currently serving, or has served, on the Editorial Boards for *Macromolecules*, *Journal of Chemical Physics*, *Journal of Polymer Science*, *Polymer Physics Edition*, *International Journal of Polymer Analysis and Characterization*, *Critical Reviews in Analytical Chemistry*, *Macromolecular Chemistry and Physics*, and *Polymer*. He has served as Chair of the Division of Polymer Physics, APS (1997-8), and as Chair of the Gordon Research Conferences on Colloidal, Macromolecular and Polyelectrolyte Solutions (1998) and Polymer Physics (2000). He served on the Council of the APS from 2001-2006, and as a Member-at-Large on the Executive Committee of the Society of Rheology from 2003-2007. He has been a visiting professor at Kyoto University, Universität Mainz, the University of California at Santa Barbara, and the University of Leeds. Since 2005 he has been Director of the NSF- supported Materials Research Science & Engineering Center at Minnesota. He has authored or co-authored over 300 papers in the field of polymer science, and advised or co-advised over 50 PhD students. His research interests center on the structure and dynamics of polymer liquids, including solutions, melts, blends, and block copolymers, with particular emphases on self-assembling systems using rheological, scattering and microscopy techniques.