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Robert Simha: Pioneer in polymer physics (1912–2008)

Robert Simha published his first two papers in 1936 and his last publication is scheduled for 2009, a remarkably long, productive career in polymer physics.

Robert received his Ph.D. at the age of 23 from the Institute of Theoretical Physics at University of Vienna for the work on colloid hydrodynamics. This was at a time when the existence of polymeric substances had just been demonstrated, and Staudinger had found experimentally a relationship between molecular weight and viscosity. For the post-doctoral stage, he joined Herman Mark's Institute at the university with its focus on polymers. There his contributions to the scientific literature began with extension of the Einstein theory of suspension viscosity to higher concentration through a consideration of binary interparticle interactions and to parabolic Poiseuille flow. With Fritz Eirich, he carried out theoretical and experimental explorations of solution and suspension viscosity, surface tension and heats of vaporization of chain molecular fluids. Mark's department depopulated in 1938 and Robert left for the USA. The work with Eirich made use of the transition state theory, recently formulated by Eyring, Polanyi and Wigner, which was put to further good use in a review chapter for the Handbuch der Katalyse, written with Herman Mark, entitled Atomphysikalische Grundlagen der Katalyse. The fee for this contribution was enough to purchase second-class passage by ship from Boulogne to New York.

Robert joined Columbia University where, as a research associate, he extended Einstein's viscosity theory to ellipsoidal solutes, and developed a general kinetic theory of chain degradation with Elliot Montroll. In 1941 he reunited with Herman Mark at the Brooklyn Polytechnic Institute, but within a year assumed his first faculty position at Howard University in Washington, DC. Subsequently, in 1945 he joined NBS as a Consultant and Coordinator of Polymer Research. During the six years at NBS, Simha worked on the quantitative theory of depolymerization with Leo Wall, experimentally confirmed by Sam Madorsky. Next, he joined New York University, where he conducted research on the statistical thermodynamics of the liquid state, as well as the viscosity of polymer solutions in good and poor solvents. Parallel with this work, Robert made an excursion into the hydrodynamics of suspensions, publishing in 1952 a highly successful viscosity-concentration relation, valid over the whole range from the Einstein limit to close packing, based on the cell model of intermolecular interactions. The theoretical equation has complex algebraic form, but only two parameters: the intrinsic viscosity and the maximum packing volume fraction. Replacing their theoretical values for monodispersed hard-sphere particles (the model used in derivation) by experimental quantities makes the theoretical relation the most successful for describing the relative viscosity of a great variety of suspensions.

In 1958, Robert moved to the University of Southern California (USC). There he reached the peak of his involvement in professional organizations, acting as head of the ACS Polymer Group, organizer and chairman of the winter Gordon Research Conference, and organizer of a series of seminars and summer courses with such lecturers as the Nobel Prize winners, Linus Pauling or Peter Debye. In 1961 he and Leszek Utracki began working on concentration, temperature, solvent quality and chain stiffness dependence of solution viscosity; a collaboration that lasted 47 years. Also at USC. Robert started collaboration with Ray Boyer on the equilibrium and non-equilibrium properties of polymer melts and glasses, general correlations between glass transition temperature, $T_{\rm g}$, and the thermal expansivity at T_g . This was followed by a series of explorations of sub-glass relaxations with Bob Haldon. However, undoubtedly the most significant development at USC was the derivation of the cell-hole theory of liquids with Thomas Somcynsky. The theory for spherical and chain molecule liquids not only correctly describes the temperature and pressure effects on specific volume (PVT), but also that of the model hole fraction, i.e., the free volume content.

In 1968, Robert Simha joined Case Western Reserve University. At Case, he created, and taught until mandatory retirement in 1983, the core course in Polymer Physics, highly popular with students, who fondly remember his lectures on chain statistics, which featured a highly realistic



Polymer Physics Symposium dinner in Old Montreal, 17 October, 2007; from the left: Ivan Otterness, Leszek Utracki, Robert Simha, Colette Lacabanne

classroom demonstration of a random walk. In addition, he continued to make ground-breaking research advances. Collaboration with Alexander Silberberg focused on the kinetics of cooperative processes in macromolecular structures. synthetic or biological. His student Robert H. Lacombe showed that these questions could be approached by detailed balancing of opposing rates and the results were examined for a spectrum of linear structures, chain lengths and sequential distributions. He continued to work on the properties of the condensed polymeric state. Anh Quach constructed a pressure dilatometer, performed careful PVT measurements on two polymer melts, and compared the results with the prediction of cell-hole theory, demonstrating for the first time the quantitative agreement. The experiments extended from high temperature to sub-Tg; comparison with the theoretical expression led to the important concept of frozen free volume content within the vitreous state. The results stimulated similar research at Case and in other laboratories, extending the success to over 50 polymers and their mixtures. In this area, Simha had wide-ranging international collaborations, e.g., with Raj K. Jain (India) extending the hole-cell theory to multicomponent systems, with Eric Nies and Alex Stroeks (The Netherlands) to phase equilibria, with Hankun Xie (China) and Chul B. Park (Canada) to gas solubility, with Leszek A. Utracki to extrusion-foaming of polymers, as well as to the interpretation of the molecular dynamics in molten and glassy polymeric nanocomposites.

Robert also provided a semi-theoretical approach to non-equilibrium properties through the use of the computed hole fraction. With John McKinney (NBS) Robert has shown that, upon cooling, the free volume partially freezes at T_{g} , with Leszek A. Utracki, the relationship between the hole fraction/free volume and viscous flow was demonstrated. Various connections between free volume and positronium annihilation lifetime spectroscopy (PALS) were studied with Profs. John McGervey, Alex M. Jamieson, Gianni Consolati and Franz H. Maurer. Another extension consisted of a theory of elastic constants of polymer glasses by Elisabeth Papazoglou, which involved the stress dependence of free volume. Finally, he analyzed the dynamics of volume relaxation, or physical aging, with John G. Curro and Richard E. Robertson, treated as a dynamics of free volume states, where the connection between the two volume quantities is given by the hole theory.

Thus, up to the last day of his life Robert Simha was an active, productive and original scientist. He was a cherished member of the polymer physics community, enlivening scientific discussions and social events with his wry sense of humor and superb knowledge of classical music. His keen insights and exceptional memory of polymer science made him a valuable resource to students and colleagues alike. His manifold contributions are bedrocks for further progress in polymer science.

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