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Institut de Génie Chimique (IGC-I) A. Polymères et Biomatériaux

Prof. Dr. **David Hunkeler***

IGC-I is principally involved in investigations on the synthesis, characterization, and application of water-soluble polymers. This includes acrylics, polysaccharides, and biopolymers with utility in environmental maintenance (water clarification), resource extraction and processing, as well as biomaterials. The primary synthetic challenges have involved the maximization of the coil size in aqueous solution as well as the optimization of the charge density along the polymer backbone. These hydrophilic macromolecules also present challenges in characterization, and we have been involved in methods development in light scattering and liquid chromatography. These analytical investigations, in addition to addressing unresolved issues such as the simultaneous characterization of polymer composition and molecular-weight distributions, also provide feedback to kinetic studies which aim to elucidate an elementary reaction scheme for various heterophase syntheses. Recently, the laboratory has also become involved in the testing of polymeric flocculants and in the formation of semi-permeable membranes. The latter, produced in microcapsular form from a complex coacervation reaction, have been evaluated as immunoisolation barriers. Of particular interest has been the development of multicomponent polyanion and polycation blends for the encapsulation of pancreatic islets and the development of bioartificial organs. It is this latter research which will be the principal theme of IGC-I over the coming years.

Biomaterials

Research to date has primarily been focused on the screening of potential polymers as semi-permeable membranes for applications in cell encapsulation. This has included an evaluation of the complexation reaction between over 1200 oppositely charged polyelectrolyte pairs. Polymer structure-function correlations have also been developed and several tens of quaternary polymer blends have been tested for biocompatibility and cell cytotoxicity. The result is a family of polymers which can serve as substitutes for the traditional lysine-alginate systems. In particular, a blend of rigid polysaccharides has been found to be an ideal media for cell suspension and to provide a relatively porous skeleton for gelation. The addition of multivalent cations and either oligomeric or macromolecular cations permits the precise control of both membrane thickness and permeability, enabling the decoupling of mass-transfer characteristics from mechanical properties. The tradeoff between rupture stability to hydrodynamic forces and molecular-weight cutoff has limited the applicability of several existing polyelectrolyte-based systems. Some of the most promising of these capsular membranes have been tested for the encapsulation of pancreatic islets and have resulted in the reversal of diabetes in NOD mice for periods exceeding three months. The *Figure* shows an example of a 'walled' capsule. Further research is under investigation on the effect of capsule size and permeability as well as the mechanism of

David Hunkeler, born in 1962, has worked at the EPFL since September 1, 1996. David obtained a Bachelors in Engineering and Management from McMaster University (Canada) in 1984 and a Ph.D. in Polymers in 1990. Between 1990 and 1996, David was an Assistant Professor of Chemical Engineering, Management of Technology and Materials Science at Vanderbilt University in Nashville, TN. He also spent one year as a visiting professor in the Department of Materials (EPFL). David's research includes polymer synthesis and characterization, encapsulation, biomaterials as well as life cycle assessment. David has authored over 50 refereed papers, edited one book and has organized five conferences. He conducts sponsored research projects with firms in the United States and has extensive research collaborations with former Eastern European countries. David teaches courses in Chemical Engineering and Process Simulation.

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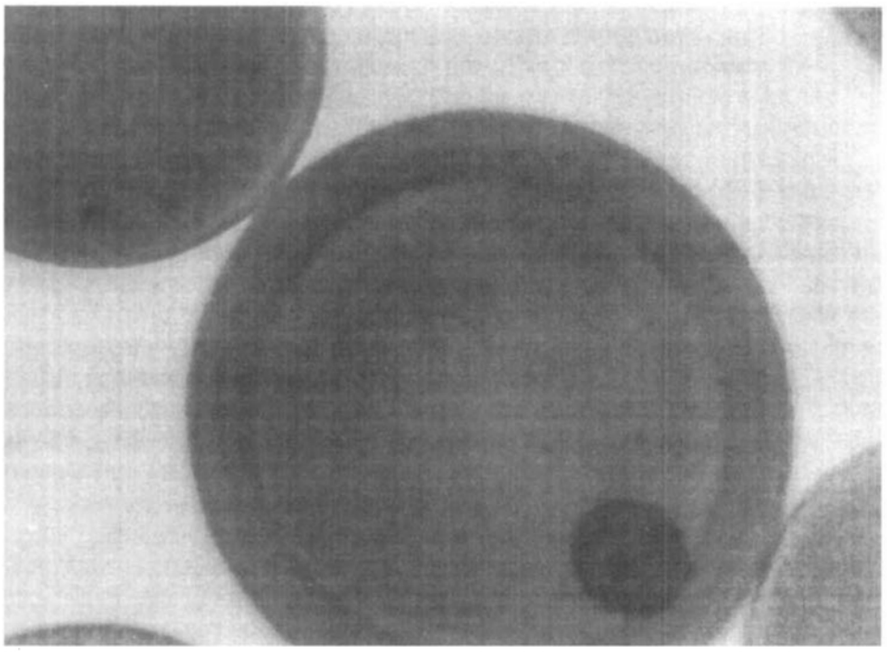

Figure. *Optical photomicrograph of an inner polysaccharide bland consisting of sodium alginate and cellulose sulfate.* Ionotropic gelatin was carried out in the presence of calcium chloride with polymethylene-co-guanidine used to control the membrane thickness and permeability. The diameter is *ca.* 650 nm. The membrane wall and islet are evident.

membrane formation and the molecular modeling of oppositely charged polyelectrolyte interactions. The design of bioreactors capable of producing pilot-scale quantities (kg/h) of microencapsulated cells is also under development.

Water-Soluble Polymers

The mechanism of the heterophase polymerization of acrylic water-soluble copolymers is under investigation. This includes water-in-oil processes such as inverse-emulsion and inverse-microemulsion as well as solvent-free or water-in-water precipitation polymerizations. The experimental projects include investigations of the stabilization and inversion of polymer latices as well as the synthesis and evaluation of novel polymeric stabilizers, individually and as components of emulsifier blends, for the production of 'non'-settling water-in-oil emulsions. Kinetic studies are also conducted to develop reactor models for the prediction of variables such as the conversion, heat generation, polymer composition, molar mass, and particle size as a function of the reaction time. The production of uniformly charged acrylamide-based polyelectrolytes through semi-batch feed strategies is also being correlated with the efficiency of quaternary-ammonium-based copolymers in applications such as potable water clarification. The development of advanced control algorithms, through the application of neural networks and fuzzy logic, is also being performed. The effect of oxygen on the free-radical mechanism is a topic of investigation.

Polymer Characterization

Analytical methods for the characterization of complex homopolymers, copolymers, and polymer blends are under development. These primarily involve the application of liquid chromatography measurements where the entropic and enthalpic separation mechanisms are balanced. By combining adsorption, precipitation, and exclusion a condition can be identified where the polymer elutes on the 'limit' of its solubility. Such limiting conditions of adsorption (LC-LCA) or limiting conditions of solubility (LC-LCS) result in a retention-independent exclusion between molar masses of several hundred to over one million daltons. This far exceeds the molar-mass cutoffs for competing methods such as 'critical conditions'. The retention is, however, sensitive to polymer composition, end group functionality and tacticity. The mechanisms of LC-LCA and LC-LCS are being elucidated and the methods are being applied to the characterization of components of polymer blends as well as the simultaneous molar mass and compositional characterization of copolymers. If successful, the micrograding LC-LCS and LC-LCA techniques will transform a standard low-cost liquid chromatograph into an NMR-like instrument. IGC-I is also involved in the development of methods for the characterization of high-molar mass water-soluble polyelectrolytes (natural and synthetic) by optical methods, and in the application of statistics to molecular parameter estimation.

The modification of silica sorbents and capillaries with water-soluble polymers is being carried out so that a new generation of stationary phases can be developed for the characterization of charged water-soluble polymers by gel-permeation chromatography and capillary zone electrophoresis.

Sustainable Development

This, somewhat smaller, component of our research has involved multinational surveys as to the present state of Life Cycle Assessment (LCA), as well as the convening of panels to formulate the role of universities in environmentally conscious manufacturing. This has resulted in the development of EcoDS, an environmentally conscious decision support tool. EcoDS is design to accommodate a needs identified in the various surveys: heightening the awareness of environmental issues within an organization at the earliest possible stage in the design process without altering the present corporate culture. Therefore, a user steps through a process to vertically streamline the LCA, followed by a qualification of the value system inherent within an organization or unit. Specific comparative evaluations are employed, and case studies are being developed for situations where liquid, solid, and gaseous emissions dominate. These include coating alternatives, fluorescent light bulbs and solvent substitution. The evaluation then proceeds fully utilizing the cost data at hand, as is preferred by most firms in comparison to point or ecounit based schemes. This financial information is complimented by a qualitative metric, which estimates the residual risk. Therefore, depending on the preferred investment and time commitment, an organization can, by selecting a suitable position on the cost-residual risk continuum, streamline their environmental decision making process to the extend desired, and perform evaluations over periods of days, weeks, or months. In the future we plan to adopt the EcoDS methodology to water treatment alternatives as well as other issues of particular relevance to the Swiss economy.