

LABORATORY OF POLYMER SEPARATION AND CHARACTERIZATION

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The origin of this laboratory goes back to December 1942, when Dr. M. Horio, at present, Emeritus Professor of Kyoto University, established his laboratory in this institute. The research field of Dr. Horio was extensively concerned with developing work on pulp and viscose rayon, and the researches contributed much to rayon industry in this country by bringing several new processes, such as those for high tenacity rayon spinning (1939), crimped rayon staple (1939), and prehydrolyzed sulphate cooking (1944) *etc.* Joining Horio Laboratory in 1954, Dr. H. Inagaki planned to solve some fundamental problems in viscose rayon industry, laying emphasis on physico-chemical properties of polymer electrolytes in solution. Papers presented in this relation have been listed in a previous commemoration issue of this Bulletin, Vol. 44, No. 6, published in 1966.

In 1960 Dr. Inagaki was appointed as professor of Kyoto University and established his own laboratory. Four years later, this laboratory was nominated as one of nineteen research sections which were set up officially in this institute according to the Ordinance No. 4 of the Ministry of Education, and was thereby designated as Laboratory of Polymer Characterization. As a result of this change in the system, Horio Laboratory was dissolved to join the aforementioned laboratory, in which Dr. Horio collaborated up to his retirement from Kyoto University in 1969 in addition to his main responsibility for the Department of Polymer Chemistry, Faculty of Engineering, Kyoto University.

Around 1963 some staffs of Laboratory of Polymer Characterization already started a series of studies on thermodynamic and conformational properties of statistical and block copolymers in solution, including the establishment of methods for copolymer characterization. During the course of these studies it was recognized that a most important information for these studies is the compositional heterogeneity of sample copolymers to be tested. As well known, however, all available methods for deducing this information at that time could not be independent of another heterogeneity, namely, the molecular weight distribution, for the thermodynamic reason. Thus, efforts were paid intensively to establish a method which allows us to deduce this information as simply and rapidly as possible.

In 1968 it was found, in collaboration with the Central Research Institute of Sekisui Chemical Industry, Co., Ltd., that thin-layer chromatography (TLC) could be well applied to the determination of compositional heterogeneities without interference of molecular weight. Since then, a variety of separation feasibilities of polymers by TLC have been demonstrated by the staffs of this laboratory, as will be seen in the publication list (classification I. 1.) given later. In view of the importance of separa-

tion science for polymers, we attempted to reorganize our research projects and changed the legal laboratory name to Laboratory of Polymer Separation and Characterization in April 1975. In the following the research projects having been finished to date and being now in progress will be described briefly.

As will be seen in the publication list, our research projects are roughly classified into three categories, namely, separation and characterization of polymers (Project I), thermodynamic and conformational properties of polymer chains (Project II), and physico-chemical properties of wool keratin and its derivatives (Project III).

Project I is concerned largely with problems of polymer separation and characterization. One of characteristics of current polymer industry is the growing interest in polymers having rather complex chain architectures or well-defined microstructures as new materials. In view that these raw materials usually contain several polymeric byproducts, we laid our emphasis on polymer separation which should be done in advance of polymer characterization. This is the reason why we started this project as the primary one. As tools for polymer separation, we are using TLC, gel permeation chromatography and various ultracentrifugation methods, and investigating, at the same time, the theoretical backgrounds of these methods. In parallel to the separation work some staffs of this laboratory have been engaged in preparative work of polymers with different chain architectures and microstructures, which were used as reference samples for separation researches. In addition TLC technique has further been applied for elucidating various polymer reactions, such as graft and block copolymerization.

Principles of the aforementioned separation techniques as well as results of polymer separation are often related to the physico-chemical properties of polymer solutions, and Project II deals rather generally with thermodynamic and conformational behavior of polymeric chains from experimental as well as theoretical aspects. Therefore this project covers problems of the excluded volume effect in homo- and co-polymeric chains in solution, and of the chain conformation of polymer electrolytes in solution. Our main interest consists, at present, in the conformational problem of block copolymer chains in solution, and the problem has been and is being examined by computer simulation and light-scattering measurement. In addition the chain microstructures of isomeric polymers were studied by some staffs of this laboratory.

Project III was started originally to elucidate the excellent property of wool fiber over other natural and synthetic fibers from an aspect of higher order structures of the component keratin molecules. To prepare chemically unmodified histological protein components from wool fiber, experimental procedures for the disruption and dissolution of wool have been investigated. During the course of this work we had an idea for utilizing kerateines, which are obtainable by reducing the disulfide bonds of wool, as a starting material to prepare a selective adsorbent-gel for heavy metal ions. Thus we are now investigating interactions of kerateines with metal ions and biologically interesting substances, possible chemical modification of kerateines to change the selectivity to ions, and practical procedures to form gel particles and films.

Last to be mentioned concerns those who contributed to our research activities by joining this laboratory as visiting scientist during this decade beginning in 1966. They are: Dr. F. Kamiyama, Sekisui Chem. Ind., Ltd. (1969-1971), Mr. K. Ban, Japan

Exlan Co., Ltd. (1971–1973), Prof. R. Kirste, Mainz University, FRG (1973), Prof. H.-J. Cantow, Freiburg University, FRG (1973), Dr. T. Ogawa, Ube Chem. Ind., Co., Ltd. (1974–1975), Dr. J. Wälchli, Bern University, Switzerland (1974–1976), Mr. T. I. Min, Pusan National University, Korea (1973–) and Dr. J. Schaller, Bern University, Switzerland (1976–). It is acknowledged with many thanks that the stay of the above foreign scientists in this laboratory was made possible by the support of either the Education Ministry or the Japan Society for Promotion of Sciences (JSPS). In 1977 it is expected that Dr. H.-G. Elias, Director of the Midland Macromolecular Institute, USA, and Prof. H. Benoit, the Centre de Recherches sur les Macromolécules, France, will join this laboratory by the support of the JSPS.

Publications

(* indicates an article published in Japanese)

I. Separation and Characterization of Polymers

I-1. Polymer Separation by TLC

1. H. Inagaki, H. Matsuda, and F. Kamiyama: Determination of Compositional Heterogeneity in Copolymers by Thin-Layer Chromatography. I. Preliminary Results for Styrene-Acrylate Copolymers, *Macromolecules*, **1**, 520 (1968).
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3. H. Inagaki: Anwendung der Dunnschicht-Chromatographie bei der Untersuchung Makromolekularer Substanzen, *Bull. Inst. Chem. Res., Kyoto Univ.*, **47**, 196 (1969).
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12. T. Miyamoto: Thin-Layer Chromatography of Polymers, *High Polymers, Japan*, **23**, 378 (1974).*
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14. F. Kamiyama and H. Inagaki: Some Fundamental Properties of Developer Solvents in Thin-Layer Chromatography Applied to Polymer Separations, *Bull. Inst. Chem. Res., Kyoto Univ.*, **52**, 393 (1974).
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17. T. I. Min, T. Miyamoto, and H. Inagaki: Thin-Layer Chromatographic Separation by the Difference in End-Group, *Bull. Inst. Chem. Res., Kyoto Univ.*, **53**, 381 (1975).

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I-2. TLC Application to Polymer Chemistry

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24. T. I. Min and H. Inagaki: Separation and Characterization of Cellulose-Styrene Graft Copolymers (II), *ibid.*, **24**, 635 (1975).*
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I-3. Gel Permeation and Column Chromatography

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I-4. Ultracentrifugation

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- I-5. Compositional Heterogeneity in Copolymer Systems (Theories)**
50. T. Kotaka, N. Donkai, and T. I. Min: Compositional Heterogeneity and Molecular Weight Distribution of Copolymer Systems. I. Simple Statistical Analysis of the Heterogeneities of Block and Graft Copolymers, *Bull. Inst. Chem. Res., Kyoto Univ.*, **52**, 332 (1974).
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II. Thermodynamic and Conformational Properties of Polymer Chains

II-1. Homopolymers in Solution

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53. H. Suzuki, T. Kotaka, and H. Inagaki: Shear-Rate Dependence of the Intrinsic Viscosity of Flexible Linear Macromolecules. II. Solvent Effect, *J. Chem. Phys.*, **51**, 1279 (1969).
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II-3. Chain Conformations

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III. Physico-Chemical Properties of Wool Keratin and Its Derivatives

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