

INAGAKI LABORATORY (September 1960~)

Head: Dr. Hiroshi Inagaki

In 1960 this laboratory was established after Dr. Inagaki was appointed professor of this University. Since 1954 he had been a member of Horio Laboratory of this Institute, and his research works made during the period 1954-1960 are included in the list of publications attached to the end of this description. In the following the research projects having been made to date and being now in progress will be mentioned briefly.

I. Polymer Electrolytes in Solution¹⁻⁸⁾

Our first concern was to treat the problem of the expansion of a charged polymer chain, which may be reflected on the intrinsic viscosity of its solution. Thus the extraneous salt concentration dependency of the intrinsic viscosity was studied using the system sodium carboxymethyl cellulose and water, and the concept of "effective charge" of polyelectrolyte was deduced from the experimental results. In this connection, the osmotic pressure of polyelectrolyte solutions was investigated, and an attempt to analyze the osmotic pressures in terms of the effective charge was made. This project has been again taken in this year to study the influence of stereospecificity upon polyelectrolytic properties using isotactic poly (2-vinyl pyridine).

II. Solution Properties of Neutral Polymers⁹⁻¹⁶⁾

The weight average molecular weight is the most important information for discussing thermodynamic properties of polymer solutions. The light scattering method has been known to be a quite promising tool for this purpose. Thus our first attention was paid to constructing a light scattering photometer which allows one to determine the molecular weight as well as the radius of gyration of solute molecule. Using the apparatus thus constructed the thermodynamic properties of polyvinyl alcohol in water were studied. At the same time, the apparent specific volume of macromolecules in solution was discussed on the basis of the statistical thermodynamical theory.

III. Application of Ultracentrifugal Methods¹⁷⁻²⁵⁾

In addition to the light scattering method, ultracentrifugal methods are investigated for determining the weight average molecular weight. The Archibld theory has been extended to thermodynamically non-ideal multicomponent systems. The validity of the derived equation has been tested experimentally using two Theta solvent systems as well as various good solvent systems of polystyrene and polymethyl

methacrylate.

IV. Excluded Volume Effect and Conformations of Stereospecific Polymers²⁶⁻³²⁾

The configurations of flexible chain polymers in solution deviate from the statistics of random flight when the long range interactions act between segments -the excluded volume effect. This effect upon the intrinsic viscosity $[\eta]$ has been experimentally studied. Recently a tentative equation for determining the unperturbed dimensions of chain polymers with $[\eta]$ and M_w relationships has been proposed, and its validity is tested experimentally. Syndiotactic and isotactic propylene polymers have been prepared and investigated of their unperturbed dimensions using our equation. A hypothesis for retention of chain conformations in solution has been proposed on the basis of experimental result. The formation mechanism of primary nuclei of dilute solution grown single crystals is studied in due course of the above investigation.

As an extension of this subject, we have been studying the behavior of a single macromolecule influenced by the excluded volume effect in the shearing flow field. Our primary concern is to find out experimentally the general feature of non-Newtonian intrinsic viscosities in regard to various molecular parameters such as solute molecular weight, solvent power etc.

V. Copolymers in Dilute Solutions³³⁻³⁸⁾

Dilute solution properties of random and block copolymers are studied. One of the purposes is to establish a physical method which enables us to determine the mode of monomer arrangement. The other one is to explain the properties as functions of the monomer composition as well as the mode of monomer arrangement. In parallel to the block polymer studies we have been studying the terminal group effect on the dilute solution properties.

VI. Rheological Properties of Polymer Solutions³⁹⁻⁴¹⁾

One of the staffs (T.K.) had been engaging in the study of rheological properties, specifically of normal stress phenomena, of polymer solutions in collaboration with the Physical Chemistry Laboratory, Dept. of Industrial Chemistry of this University. A summary of the study will appear in Ref. 41.

VII. Gel Permeation Chromatography

Columns containing crosslinked polystyrene gels prepared in the presence of diluents have been found to be capable of fractionating polymeric samples in organic solvents (Moore: 1964). We are studying now preparation of a new type of the gel in order to improve the mechanical stability of gel, and the fractionation mechanism.

Publications

(* indicates an article published in Japanese)

1. H. Inagaki, H. Sakurai and T. Oda: Association of Cations with Anionic Polyelectrolytes in Dilute Solution, *Bull. Inst. Chem. Res., Kyoto Univ.*, **33**, 257-264 (1955).
2. H. Inagaki, H. Sakurai and T. Sasaki: Distinct Maximum Appearing in the Viscosity Curve of Dilute Polyelectrolyte Solutions, *ibid.*, **34**, 74-86 (1956).
3. H. Inagaki: On Concentrated Solutions of Polyelectrolytes. I. Spinnability and Non-Newtonian Viscosity, *J. Colloid Sci.*, **11**, 226-236 (1956).
4. H. Inagaki and T. Oda: The Second Virial Coefficient in Polyelectrolyte Solutions with Extraneous Salts, *Makromolekulare Chemie*, **21**, 1-12, (1956).
5. H. Inagaki, S. Hotta and M. Hirami: Further Study on the Second Virial Coefficient in Polyelectrolyte Solutions with Extraneous Salts, *ibid.*, **23**, 1-15 (1957).
6. H. Inagaki: Note on the Donnan Membrane Equilibrium in Polymer Salt Solutions with Extraneous Salts, *Bull. Chem. Soc. Japan*, **30**, 115-117 (1957).
7. H. Inagaki and M. Hirami: Über den osmotischen Druck von Polyelektrolytlösungen mit Zusatzsalz, *Z. Elektrochem.* **63**, 419-427 (1959).
8. H. Inagaki and A. Teramoto: Das scheinbare spezifische Volumen von Fadenmolekularen Polyelektrolyten in Lösung, *Makromolekulare Chem.*, **47**, 185-200 (1961).
9. M. Horio, U. Imamura and H. Inagaki: A Study of Light Scattering on Xylan, *TAPPI*, **38**, 216-220 (1955).
10. H. Inagaki, Y. Tachibana and S. Kozawa: Photoelectric Light Scattering Photometer, *Shimadzu Rev.*, **13**, 55-60 (1956).*
11. H. Inagaki, Y. Takeo, T. Oyama, K. Kawahara, K. Matsunami and E. Aoki: Light Scattering Measurements in Polymer Solutions, *Nippon Kagaku Zasshi*, **78**, 676-685 (1957)*.
12. G.V. Schulz, H. Inagaki and R. Kirste: Zur phänomenologischen und statistischen Thermodynamik von Polymerlösungen, *Z. Physik. Chem. (N.F.)* **24**, 390-404 (1960).
13. Y. Kobatake and H. Inagaki: Eine thermodynamische Theorie über das scheinbare spezifische Volumen von Hochpolymeren in Lösung, *Makromolekulare Chem.*, **40**, 118-125 (1960).
14. H. Inagaki: The Apparent Specific Volume of Polymers in Solution, *Bull. Inst. Chem. Res., Kyoto Univ.*, **40**, 364-373 (1962).
15. T. Matsuo and H. Inagaki: Über den Lösungszustand des Polyvinylalkohols in Wasser. I. Metastabiler Zustand der Lösung, *Makromolekulare Chem.*, **53**, 130-144 (1962).
16. T. Matsuo and H. Inagaki: Über den Lösungszustand des Polyvinylalkohols in Wasser. II. Molekulargewichtsbestimmung durch Streulichtmessung, *ibid.*, **55**, 150-166 (1962).
17. H. Inagaki and S. Okamoto: A New Revolution Counter for Ultracentrifuges, *Bull. Inst. Chem. Res., Kyoto Univ.*, **33**, 233-236 (1955).
18. H. Fujita, H. Inakgai, T. Kotaka and H. Utiyama: Application of the Archibald Ultracentrifugal Method for the Study of Dilute Polymer Solutions. I. Theory and Some Preliminary Data on the System Polystyrene-Methyl Ethyl Ketone. *J. Phys. Chem.*, **66**, 4-10 (1962).
19. H. Inagaki, S. Kawai and A. Nakazawa: Application of the Archibald Ultracentrifugal Method for the Study of Dilute Polymer Solutions. II. Two Solution Systems near the Flory Temperature, *J. Polymer Sci.*, **1A**, 3303-3315 (1963).
20. H. Inagaki: Einige Bemerkungen über die Molekulargewichtsbestimmung nach dem Archibald-Verfahren unter besonderer Berücksichtigung der Arbeit von W. Scholtan und H. Marzolph, *Makromolekulare Chem.*, **64**, 215-218 (1963).
21. H. Inagaki and S. Kawai: Anwendung der Ultrazentrifugen-Methode von Archibald bei der Untersuchung verdünnter Polymerlösung. IV. Molekulargewichtsbestimmung von Polystyrol und Polymethylmethacrylat in Butanon, *Makromolekulare Chem.*, **79**, 42-53 (1964).
22. T. Kotaka and H. Inagaki: The Archibald Ultracentrifugation Method in the Study of Macromolecules, *Bull. Inst. Chem. Res., Kyoto Univ.*, **42**, 176-203 (1964).
23. H. Inagaki, A. Nakazawa and T. Kotaka: Temperature Dependence of the Second Virial Coefficient of Bovine Serum Albumins by the Archibald Ultracentrifugation Method, *Bull. Inst. Chem. Res., Kyoto Univ.*, **43**, 135-148 (1965).

24. T. Kotaka, N. Donkai and H. Inagaki: The Archibald Ultracentrifugation Method in Binary Solvent Systems, *Bull. Inst. Chem. Res., Kyoto Univ.*, **44**, 192–201 (1966).
25. A. Nakazawa and H. Inagaki: Effect of Solute Polymolecularity on the Archibald Sedimentation Plot, *Nippon Kagaku Zasshi*, **87**, 1049–1052 (1966).*
26. H. Inagaki, H. Hayashi and T. Matsuo: Die ungestörte Knäueldimension von Polyacrylnitril unter Berücksichtigung des Einflusses der Polymerisationstemperatur, *Makromolekulare Chem.*, **84**, 80–92 (1964).
27. H. Inagaki, H. Suzuki, M. Fujii and T. Matsuo: Note on Experimental Tests of Theories for the Excluded Volume Effect in Polymer Coils, *J. Phys. Chem.*, **70**, 1718–1726 (1966).
28. H. Inagaki, H. Suzuki and M. Kurata: A Semi-Empirical Equation for Estimating Unperturbed Dimensions of Chain Polymers from Intrinsic Viscosity Data, *J. Polymer Sci.*, C-series in press.
29. A. Nakazawa, T. Matsuo and H. Inagaki: Unperturbed Dimension and Conformation of Polyvinyl Chloride Chain in Solution, *Bull. Inst. Chem. Res., Kyoto Univ.*, **44**, 192–200 (1966).
30. H. Inagaki, T. Miyamoto, and S. Ohta: The Unperturbed Dimensions of Polypropylene and Polyethylene, *J. Phys. Chem.*, **70**, 3420–3431 (1966).
31. H. Inagaki and T. Miyamoto: Einkristalle von Syndiotaktischem Polypropylen aus Verdünnter Lösung, *Makromolekulare Chem.*, **987**, 267–271 (1966).
32. T. Kotaka, H. Suzuki, and H. Inagaki: Shear Rate Dependence of the Intrinsic Viscosity of Flexible Linear Macromolecules, *J. Chem. Physics*, **45**, 2770–2773 (1966).
33. H. Inagaki: Das scheinbare spezifische Volumen von Copolymeren in Lösung, *Makromolekulare Chem.*, **75**, 217–221 (1964).
34. H. Inagaki and M. Tanaka: Endgruppeneffekt des Knäuelmolekuls auf Lösungseigenschaften. I. Polyathylenglykol mit ionisierbaren Endgruppen, *Makromolekulare Chem.*, **74**, 145–157 (1964).
35. H. Inagaki: Intra-Chain Interaction and Molecular Shape of Block Copolymer in Dilute Solution, *Makromolekulare Chem.*, **86**, 289–293 (1965).
36. H. Inagaki and T. Miyamoto: Preparation of Block Copolymer of A-B-A Type and its Behavior in Dilute Solution, *ibid.*, **87**, 166–179 (1965).
37. H. Ohnuma, K. Igi, T. Kotaka, and H. Inagaki: The Terminal Group Effect on Solution Properties of Linear Macromolecules: Preliminary Results, *Bull. Inst. Chem. Res., Kyoto Univ.*, **44**, 123–134 (1966).
38. T. Kotaka, H. Ohnuma and Y. Murakami: The Theta Condition for Random and Block Copolymers of Styrene and Methyl Methacrylate, *J. Phys. Chem.*, **70**, 4099 (1966).
39. T. Kotaka and K. Osaki: Dynamic Viscoelastic Behavior of Concentrated Polymer Solutions. (I), *Bull. Inst. Chem. Res., Kyoto Univ.*, **39**, 331–340 (1961).
40. T. Kotaka, M. Kurata and M. Tamura: Non-Newtonian Flow and Normal Stress Phenomena in Solutions of Polystyrene in Toluene, *Rheologica Acta*, **2**, 179 (1962).
41. T. Kotaka and K. Osaki: On Normal Stress, Non-Newtonian Flow and Dynamic Mechanical Behavior of Polymer Solutions, *J. Polymer Sci.*, C-series, (1966) in press.
42. T. Kotaka and R.L. Baldwin: Effects of Nitrous Acid on the dAT Copolymer as a Template for DNA Polymerase, *J. Mol. Biol.* **9**, 323 (1964).