Santi R. Palit

SANTI RANJAN PALIT

(1912—1981)

Elected Fellow 1953

Birth, Parentage and Childhood

Santi Ranjan Palit was born in Calcutta on March 24, 1912. His ancestral home was in Gabdhan in the District of Barisal, now in Bangladesh. His father, Naranarayan Palit, came to Calcutta after passing Entrance examination from Barisal. In Calcutta, he got a job as a cashier in the estate of the then renowned Sil family. He was a very kind and honest person. Santi Ranjan's mother Srimati Kusum Kumari had a great yearning for learning. She was herself a brilliant student in her career, though she had to give up formal studies at a very early stage. She continued, throughout her life, to improve her knowledge through self-education. Santi Ranjan was the only son amongst four sisters. He was greatly influenced by his mother, and it is by her insistence he could continue his higher education. This was particularly so since his father wanted him to take up a job after passing matriculation.

Santi Ranjan grew up in an old-world atmosphere, so vividly and interestingly described in the book *Mahasthabir Jatak* by the noted Bengali literary Premankur Atarthi. The Atarthis were their next-door neighbour. Santi Ranjan spent his childhood and boyhood days in a playful atmosphere amongst other boys of their joint family.

Education

Santi Ranjan's early education was in Calcutta Training Academy which was at a stone's throw from their house. The Academy had few permanent teachers. There was one outstanding mathematics teacher, Sri Rabindra Nath Majhi, whose excellent logical exposition of the subject made a lasting impression on young Palit. However, he was attracted to science mainly by reading a couple of Bengali books on science written by the famous science book writer Acharyya Ramendra Sunder Tribedi.

Santi Ranjan passed Matriculation in 1927 from the Academy with star and letter marks in four subjects. He passed ISc from Vidyasagar College in first division. After passing ISc examination, Santi Ranjan, due to illness of his father, was compelled to accept a job in the estate of Sil family. It was at the mother's insistence and encouragement that Santi Ranjan gave up the job and got admitted in the BSc (Honours) in the Scottish Church

College. He stood first class first in BSc with Chemistry (Honours) in 1931. He repeated the same feat in the MSc examination in 1933. After this he faced the bleak prospect of joblessness. That was a period of strong Swadeshi movement and his mother, being an ardent disciple of Gandhiji, did not like the idea that her only son should sit for any kind of public service examination in order to become a slave of British ruler. She had earlier advised her son not to take his degree from the hands of the British Chancellor at the Convocation. To his mother tea was a taboo as it was grown by British planters and Palit, following his mother's example, never took tea in his life.

Professional Career

Santi Ranjan had to spend two long years after MSc on job hunting. Dr Shyama Prasad Mukherjee, the then Vice Chancellor of Calcutta University, took interest in him and made a research scholarship available to him. After toiling for one year at the Physical Chemistry Department of the Science College, he joined as a lecturer in Vidyasagar College and taught there for two full sessions. He was a very successful teacher and students, both of Honours and Pass courses, felt greatly attracted towards his lectures and eagerly waited for attending his classes. During this time, he authored the book *Elementary Physical Chemistry* which became immensely popular amongst students and teachers alike.

In July 1938, Santi Ranjan joined the Lac Research Institute at Namkum, Ranchi, as a Research Assistant under Dr HK Sen. In the interview for the post held at Simla, he pointed out to Sir SS Bhatnagar, a leading member of the selection committee, that some of the statements made by the latter are factually incorrect. An impressed Bhatnagar not only selected him for the post but took a fatherly interest in him for the rest of his life. His first paper from Lac Research Institute on cosolvency of shellac was well appreciated by the scrutinising committee. Since then he was given free hand in his research work, which earned him the Premchand Roychand Scholarship (PRS), a coveted award at that time and also the DSc degree of the Calcutta University. In 1943, after his father's sudden death he resigned his job and left the place.

His work on cosolvency drew the attention of Professor McBain who invited Santi Ranjan to join him at Stanford University, California. He joined Stanford University in early 1945 and put himself, heart and soul, in the researches on consolvency and solubilization. He published over half a dozen papers in a span of little over one year's work. The university wanted to give him a Doctorate degree but Santi Ranjan did not like to add any additional Doctorate degree to the DSc degree he already had from his *Alma Mater*. In 1946 he came to New York and joined Professor Herman Mark at the Polytechnic Institute of Brooklyn where he was introduced to the then rapidly growing field of polymer science. He also worked for a few months in an industrial research laboratory under Dr Valko, a friend of Dr Mark, to gain experience. He returned to India a couple of months before Independence.

On his return to India, he met Professor MN Saha and won his life long trust and support. Dr Saha was then the President of the Indian Association for the Cultivation of Science, Calcutta, and appointed him as the Professor and Head of the Department of Physical Chemistry in the same institute in 1947. It was the year of Independence and there was a spirit of intense nationalism and an urge to forge ahead all around. It was rightly recognized that the development of science and technology was an integral part of the progress of a nation. The Association under the initiative of Professor MN Saha undertook a programme of expansion of its activities and drew up a comprehensive and integrated plan of research. Santi Ranjan joined the Association in this crucial period and actively engaged himself with all his vigour and enthusiasm in the development plan of the Association. He made significant contributions in many areas of physical chemistry as well as polymer chemistry. His contributions in the field of polymer chemistry are of lasting importance and he is justifiably regarded as the father of polymer research in India. Professor Herman Mark of USA, the doyen of polymer scientists of the world, described him as the "leading polymer scientist in India and in fact, the most prominent representative of this discipline in the Far East, a recognized member of the small group of leading polymer scientists in the entire world".

During the last ten years of his life, Palit was deeply engrossed in his researches on non-Faradaic electrolysis. He continued to work in the filed even after his formal retirement from the Association, after serving for 30 years, in December 1975. He was made an Emeritus Professor, the position he held till his death. He was very fond of the Association and never thought of leaving it in spite of getting lucrative offers from other research establishments. His varied interests and prolific research activities are evident from the fact that he published more than 350 research papers and successfully guided more than 80 research students for their PhD degrees.

Santi Ranjan was a successful writer. Apart from his well-known book *Elementary Physical Chemistry*, he published a Bengali version of the same book and coauthored a text book on *Practical Physical Chemistry*. The prominent features of these books are their clarity of expression and lucid style combined with scientific reasonings. These books have been very popular with chemistry students and teachers alike.

Contributions to New Knowledge

Palit made notable contributions in the field of cosolvency, non-aqueous titration, solubilization, dipole moment, thermodynamics of liquid mixtures, proton transfer reaction in aprotic media, polymer chemistry and electrolysis. He was remarkably active till the last days of his life. When he was nearing

60, he came out with his important findings on electrolysis which was subsequently referred to as non-Faradaic electrolysis. This speaks of his keen interest in research and boldness of thought and conviction without which pioneering contribution would not have been possible. Some of his important contributions, particularly of the pioneering type, are discussed in following pages.

Cosolvency

Palit made his mark as a researcher while a beginner in scientific research in Lac Research Institute at Namkum, Ranchi. In his first paper on physical chemistry of resin solutions (1940) he elucidated the cause underlying the anomalous solubility of resins such as shellac, copal etc. in organic solvents. He demonstrated that the anomaly was due to the presence of small quantities of polar impurities, very often moisture, present in the resin or in the solvent or in both. He showed, for example, that pure dry shellac is not soluble in dry acetone, it is not soluble in water either but soluble in a mixture of the two. Of course, the phenomenon of dissolution of a substance in a mixture of two liquids (none of which are solvents for the substance) called cosolvency was known at that time ; the solution of nitrocellulose in a mixture of alcohol and ether being one classical example. The anomalous solubility behaviour of resins baffled researchers and technologists working with varnishes and lacquers. Palit's work removed much of the confusion then prevailing in the technological world of varnishes. Subsequently, Palit published a number of papers on the physical chemistry of resin solutions. He proved that contrary to the widely held notion, the solutions of shellac in moist acetone or in similar other solvents are true solutions and not colloidal ones.

Until Palit showed otherwise, the cosolvency was considered typical for resinous or high molecular weight substances. Palit showed (1942) that such simple well characterized substances as soaps are best dissolved in cosolvent mixtures. He discovered that mixtures of two liquids, one being glycolic type (called G-solvent) and the other being any organic solvent which can dissolve hydrocarbons e.g. higher alcohols, chlorinated hydrocarbons etc. (called H-solvent), have better solvent power for soaps than any of the pure solvents alone. These solvent mixtures are called G-H solvents for brevity. From an exhaustive study of the cosolvency for soaps Palit satisfactorily explained the cosolvent action of G-H mixtures. The glycol forms hydrogen bonds with the COO^- group of the soap, the monohydric alcohol fails to interact as effectively as the glycols. He attributed this to the greater acidity and therefore greater H-bonding ability of glycols than monohydric alcohols. This property of glycol results from the inductive effect of one electronegative OH groups on another. He further showed that not only soaps but also inorganic salts having the group XOO^- where X is any non-metallic element e.g. nitrites, hypophosphites etc. have very high solubility in glycols owing to the glycolic interaction. His work on cosolvency was well recognized in the contemporary literature. A pionear as he was in nonaqueous titrimetry, it is no wonder that he developed a method for the direct volumetric analysis of soaps soon after he found the powerful solvent action of G-H mixtures.

Palit's interest on cosolvency remained alive throughout his career. In later life when he established himself as a distinguished researcher in polymer chemistry he studied cosolvency in the field of polymers as well. Palit and coworkers (1973) discovered several cosolvent systems for dissolution of ethylene low polymers at ambient temperatures. It is known that polyethylene, a polymer of great commercial importance, can be taken into solution only with difficulty. For example, it can be dissolved in xylene only at high temperature. The finding that low molecular weight polyethylene can be taken into solution at ambient temperatures evoked considerable interest. Deb and Palit (1969) was able to predict the cosolvent composition of best solvent power using the 'single liquid approximation' model of Scott as a starting point.

Non-Aqueous Titration

Palit was a pioneer in the field of non-aqueous titration. Soon after his discovery of the powerful cosolvent action of G-H mixtures for soaps, Palit (1946) showed that soaps can be satisfactorily titrated by strong acids in these glycolic solvents. He found that weak bases such as ammonia or alkyl amine shows a strong base titration curve in glycol and bases such as sodium acetate which are too weak to be titrated satisfactorily by strong acidsin water, can be titrated directly with sharp end points by strong acids in glycolic solvents. Palit pointed out that since glycol is an acidic or protogenic solvent, one would expect it, according to the Lowry-Bronsted theory, to augment the apparent basic power of a base and beyond a certain strength make all bases appear strong. Palit's method finds place in standard analytical texts dealing with the analysis of the substances mentioned above. The analytical texts also recommend the use of acetic acid as a solvent for titration of weak bases. The G-H solvents do not usually yield as sharp end points as acetic acid but they are generally better solvents and the mixtures can be varied to dissolve a wide range of materials. Furthermore, the G-H solvents usually permit better differentiation of multiple bases present in the same mixture; in acetic acid media only one end point including all bases in the sample is obtained. Palit's method was extended by many workers notably Siggia in USA and Kreshkov in USSR. Palit also found that inorganic salts such as mercuric acetate, can be satisfactorily titrated in glycolic solvents. Based on this observation, Palit and Somayajulu (1955) developed a method for the volumetric determination of mercury. Palit also co-authored a monograph on the subject entitled *Non-Aqueous Titration* which was published by Indian Association for the Cultivation of Science in 1954. The work turned out to be an important treatise on the subject and soon found translated into Russian language.

Solubilization

Palit's work in the field of solubilization (1949-1954) was devoted towards understanding the mechanism of the process of solubilization of water in organic solvents. Palit *et al* supported the mechanism proposed by Winsor in this respect. They further showed (1959) that the process of solubilization involves a number of phase transitions and very often liquid crystalline phases are found. The peculiar electro-optical properties of these liquid crystals were studied and interpreted in terms of Winsor's theory. They also constructed the ternary phase diagram of the system lauryldimethylbutyl-ammonium bromide + chlorobenzene + water which helped in elucidating the process of solubilization. Palit's works in this area were extensively quoted in a number of books.

Dipole Moment Computation

For the computation of true polarization of solute from solution data, many workers proposed a number of methods all of which were based on Debye's classical equation. Of these the most important ones were those proposed by Guggenheim (1949) and Smith (1950). However, these methods were based on several assumptions which are questionable. Palit and coworkers (1951) pointed out that since polarization is expressed in volume units, the rational approach to the problem of computation of polarization without involving any assumption should use the concept of partial specific polarization in mixtures. Based on this concept, they developed a theoretically rigorous method for the computation of polarization at infinite dilution and hence the dipole moment-either graphically or analytically. The method developed by them is a quite general one. It does not use Debye's equation. Furthermore, unlike the other methods the computation procedure using Palit's method is much simpler and it produces reliable results. Also, the method enables to determine the change in polarization for a component in a solution with change in composition which, in turn, helps to throw light on their mutual interaction. However, almost simultaneously Guggenheim (1951), using an elaborate mathematical procedure, developed a new equation which was much simpler than the equations proposed by him alongwith Smith (mentioned earlier in this section). Immediately afterwards Palit (1952) showed that his partial specific polarization approach leads to a more complete equation for dipole moment computation, fairly good approximate of which is only represented by Guggenheim's equation.

Polymer Chemistry

Palit was a distinguished pioneer in the field of polymer science. In fact, researches in the field of polymer science in India started in his laboratory. He made very significant contributions in areas such as (*a*) Chain transfer reaction, (*b*) Kinetics of polymerization, (*c*) Aqueous polymerization and (*d*) Determination of endgroups in polymers vis-a-vis mechanism of polymerization. The most significant work for which Palit will be remembered by generations of polymer chemists is his discovery (1959-60) of two simple techniques called the dye techniques for the determination of end groups in high polymers. The highlights of his research in this and other areas mentioned above are presented in the following subsections.

Chain Transfer Reaction

Palit and coworkers determined reliable chain transfer constants for various solvents in radical polymerization of methyl methacrylate, vinyl acetate and acrylonitrite with a critical examination of the underlying theory and interpretation of data. These data have been quoted extensively in literature and in polymer texts.

Kinetics of Polymerization

Palit and coworkers determined kinetic constants for homogeneous free radical polymerization using initiators such as H_2O_2, persulfate, organic peroxides, azonitrites etc. Palit (1955) correlated theoretically the cross termination ratio in copolymerization reactions, the so-called ϕ factor, with rate and degree of polymerization. He and his coworkers then proceeded to determine with success the values of ϕ experimentally using the correlation mentioned above for various pairs of monomers.

Aqueous Polymerization

Palit and coworkers made extensive investigations on aqueous polymerization and demonstrated that the stability of the latex formed in heterogeneous aqueous polymerization is a very important factor in their kinetics.

Endgroups in High Polymers

As has been stated before, the most significant contribution of Palit in the field of polymer chemistry is his discovery (1959-60) of two simple techniques called the dye techniques for the determination of endgroups in high polymers. Endgroups constitute a very minute part of the polymer molecule. They are not amenable to ordinary chemical or physical methods of analysis. Special methods such as the tracer method are invariably used whenever information on endgroup content of a polymer is desired. However, the tracer method requires radioactive labelled initiators and is therefore hazardous, expensive, requiring special chemicals and equipments. Also, the method is often incapable of giving results specific to the actual nature of functional group.

In contrast, the dye techniques introduced by Palit is simple, easy to operate requiring only common place equipments and chemicals. These methods take advantage of the fact that the molar extinction coefficients of dyes in general, are very high being of the order of 10^5 litre $mole^{-1}$ cm^{-1}. Therefore, tagging polymer endgroups with dyes or reacting them with dyes to produce colour reactions would make the endgroups amenable to test. Two techniques viz., the dye partition and the dye interaction, were developed by Palit for the above mentioned purpose. These techniques can however determine only acidic or basic groups or their salts such as sulphate, sulfonate, carboxyl, amines etc. or groups which can be so transformed such as OH, halogen bearing groups. Hydroxyl groups can be transformed to COOH or SO_3H groups by suitable chemical treatment, while halogen bearing groups can be transformed to quaternary ammonium salts by quaternization reactions.

The principle of the dye partition test lies in the fact that dyes could be extracted from aqueous solutions into organic diluents by polymers which have a charge opposite to that of the dye. For example, the extraction process involving a cationic dye and a polymer with anionic endgroups may be represented as follows

$$\sim SO_3^- M^+ (org) + Dye^+ (water) \rightleftharpoons \sim SO_3^- Dye^+ (org) + M^+ (water)$$

Where $\sim SO_3^- M^+$ represents a polymer with sulfonate endgroup being present as ion pair (M^+ being H^+ ion or any univalent cation). The organic phase containing the polymer under test, thus, becomes coloured. The measurement of the colour intensity of the organic phase provides an estimate of the concentration of the ionic end function of the polymer. For the determination of strong acid endgroups, such as sulfonate or sulfate, methylene blue is the dye used with success. For weak acid groups such as COOH, pinacyanol in alkaline solutions is the recommended dye. Similarly, for the determination of cationic end function, such as those derived from amine groups following protonation or quaternary ammonium groups, an acidic solution of the anionic dye, disulfine blue VN 150 is used. Proper choice of organic diluents, pH, dye etc. provides selectivity in analysis. Subsequently, the dye extraction process was analysed theoretically by Mandal and Palit (1971) and the theoretical treatment identified the correct procedure to be followed for quantitative estimation.

The dye interaction method uses the colour reaction between acidic, basic or even salt groups present in non-polar polymers with dyes having suitable functional groups. For example, for the estimation of basic functions an acidic dye of the xanthene class such as eosin, rose bengal, erythrosin etc. is used in benzene solution. Similarly, for the estimation of acidific functions dyes of the rhodamine class were used. In view of the fact that the colour reaction is complex and not necessarily complete, the method found limited quantitative applicability.

The importance of endgroup analysis lies in the fact that information about its chemical nature and content provide important clues to the mechanism of polymerization particularly of initiation, termination and chain transfer reactions. The free radicals generated in initiation reactions are trapped as polymer endgroups. Therefore, identification of endgroups provides important information about the transient free radical intermediate which are generated in many redox reactions when the latter are used to initiate polymerization. Using the endgroup analysis method Palit and coworkers unraveled the mechanisms of many redox reactions. The results have been summarized in a review (*Rev. Macromol. Sci. Macromol. Chem. C* 2, 225, 1968). For example, it was proved that the following redox reactions, among others, produce the free radical intermediates indicated in the reactions

$$Ti^{3+} + NH_2OH + H^+ \longrightarrow Ti^{4+} + \dot{N}H_2 + H_2O$$

$$Fe^{3+} + S = C(NH_2)_2 \longrightarrow Fe^{2+} + \dot{S}C(=NH)NH_2 + H^+$$

$$RNHCL + Fe^{2+} \longrightarrow FeCl^{2+} + R\dot{N}H$$

$$X_2 + NH_3 \longrightarrow NH_3^+ + X^- + \dot{X}$$

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^- + \dot{O}H$$

$$S_2O_8^{2-} + S_xO_y^{n-} \longrightarrow \dot{S}O^- + \dot{S}_xO_y^{(n-1)-} + SO_4^{2-}$$

$$(S_xO_y^{n-} = SO_3^{2-}, S_2O_3^{2-}, HSO_3^-, SO_2O_4^{2-}, \dot{S}_2O_5^{2-} \text{ etc.})$$

$$S_2O_8^{2-} + M_n^+ \longrightarrow \dot{S}O_4^- + M^{(n+1)+}$$

$$MnO_4^- + H_2C_2O_4 \longrightarrow \dot{O}OCCOOH + \text{products}$$

$$MnO_4^- + \text{hydroxy acids} \longrightarrow \text{COOH and OH bearing radicals}$$

$$MnO_4^- + \text{EDTA} \longrightarrow \text{COOH bearing radicals}$$

Miscellaneous

Among his other works in the field of polymer science, mention may be made of the following. Based on Eyring's rate theory and 'hole' theory of liquids, Palit (1955) deduced an equation correlating the intrinsic viscosity with molecular weights of polymer. The equation was found to satisfy experimental data for many polymers. The Mark-Howink equation was shown to be a special case of this equation.

Non-Faradaic Electrolysis

Palit (1967) made the interesting observation that electrolysis of dilute solutions at low current density produces results not compatible with those expected from Faraday's laws and Faraday-Hittorf mechanism. He identified three basic non-Faradaic features or anomalies characteristic of such electrolysis of solutions which would normally liberate hydrogen at cathode and oxygen at anode. These are :

(a) Volume deficit——The volume of gas liberated at both the cathode and anode are less than that predicted by Faraday's laws.

(b) Ratio anomaly——The volume ratio of cathode gas to anode gas is greater than 2. Faraday's laws predicts that the said ratio should be 2.

(c) Co-liberation——The cathode gas as well as the anode gas is individually an explosive mixture of hydrogen and oxygen.

All these anomalies have been found to be the general electrochemical behaviour of practically all electrolytes at low concentrations under conditions of electrolysis at low current densities. He also observed that with increase of current and/or concentrations, all the anomalies tend to disappear and the results become more and more Faradaic. Palit gave lecture demonstrations of his experiments in various laboratories of the country and of the West. Editor of *Chemistry* saw him conducting the demonstration at Guilford College, USA and published Palit's work in *Chemistry* Vol 48, 16 (1975) inviting comments from the readers.

Aside from the observations mentioned above, Palit came across a number of other interesting observations—some of these are :

(a) Intermediate zone (IZ) gas liberation—Palit found that gases (hydrogen and oxygen) are liberated not only at the electrodes but also in the zones intermediate between the electrodes in suitably designed cells.

(b) Chemical reaction in IZ—Under anomalous electrolysis conditions substances such as benzene, chlorobenzene, nitrobenzene etc. if placed in the IZ get transformed into a brown coloured complex mixture containing, among others, polyphenols. He also observed oxidation and reduction of dissolved nitrogen to HNO_3 and NH_3 respectively.

(c) Migration of electroneutral molecules from IZ.

(d) Apparent migration of anion to cathode and cation to anode

(e) Depletion in the IZ (anti-Hittorf phenomenon).

Till the last days of his life Palit carried out investigations on non-Faradaic electrolysis and came across one anomaly after another. From the detailed investigations he proposed mechanisms for the anomalous electrolysis. He also carried out investigations on galvanoluminescence. In particular, he studied the chemical reactions occurring at a luminescent cathode and the non-Faradaic nature of these reactions.

Honours

The scientific contributions of Professor Palit earned him recognition from various quarters both India and abroad. He was a Fellow of the National Academy of Sciences of India, a Fellow of the Indian National Science Academy and a Fellow of the Royal Institute of Chemistry (London). He was elected President of the Chemistry Section of the Indian Science Congress (1958), and was a Vice-President of the Indan Chemical Society (1973-74). He worked as a guest Professor at the Fritz Haber Institute der Max Planck Gesellschaft, Berlin (1965-66), Visiting Professor at the University of Florida, Gainesville, Florida (1966) and at the University of Science and Technology, Kumasi, Ghana (1976-77).

Marriage and Reminiscences of Personal Life

Santi Ranjan was married in 1935 to Srimati Ikon Basu, the daughter of Sri Hemanta Kumar Basu of Barisal. He was only 23 at that time. A short while after that his father became almost blind and he had to bear the responsibility of the whole family. His first daughter was born in 1938. After that he had another daughter and three sons born to him. The sons and daughters have all been married and all the sons and sons-in-law are well established in life.

He was a fine speaker and his lectures whether on scientific subjects or other fields were always instructive and stimulating.

He was a keen sportsman and took everything in life in a sporting manner. He was a lively conversationalist capable of quoting frequently from English, Bengali and Sanskrit literature. It was always a delight to listen to him. One never felt dull in his presence. He was also a great lover of music. He was a kind and benevolent person, and admired and respected by students and colleagues. After his death his students and admirers raised funds amounting about Rs. 80,000/- to institute a biennial All India Award of Rs. 10,000/- in his name to be given to outstanding researchers in the field of physical/polymer chemistry. The award is given by Indian Association for the Cultivation of Science from 1985.

He was active till the last days of his life. Shortly before his death he visited many universities and laboratories of the west and gave lecture demonstrations on the subject of his latest interest; 'Non-Faradaic Electrolysis'. Soon after his return from this successful lecture tour abroad he had a heart attack, from which he did not recover. He breathed his last on September 13, 1981, leaving behind his wife, three sons, two daughters, many grand children and his numerous students, friends and admirers to mourn his loss.

AK Chaudhuri
BM Mandal

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