

## NIKOLA KALLAY – *curriculum vitae*

Professor Nikola Kallay was born in Zagreb (Croatia) on 5 September 1942. He attended school in Zagreb. In 1967 he graduated with a B. Sc. in chemical engineering from the Faculty of Chemical Technology, University of Zagreb. In 1969 he gained employment at the Institute of Physical Chemistry of the University of Zagreb (later the Institute of Chemistry) as a research assistant in the team of Professor Božo Težak, and in 1978, after the Institute's incorporation into the University, he became a member of the Laboratory of Physical Chemistry, Department of Chemistry, Faculty of Science, University of Zagreb. From the same faculty he obtained his M. Sc. and D. Sc. degrees in chemistry in 1972 and 1973, respectively, writing both theses on the physical chemistry of colloidal systems. In 1978 he received his Dr. habil. degree in physical chemistry. He was promoted to the position of research associate and titular assistant professor in 1978, senior research associate in 1982, research advisor in 1984, and full professor in 1988. In 1996 he was elected as tenured full professor.

In 1977 Professor Kallay served as the director of the Institute of Chemistry, University of Zagreb. During his employment at the Faculty of Science he has served as vice dean (1982), head of the Laboratory of Physical Chemistry (1999), and chairman of the Department of Chemistry (2001–2005). In 2007, he was elected head of the Laboratory of Physical Chemistry. In addition, he is currently also the head of the postgraduate program in physical chemistry.

Professor Kallay is active in science and education. He does scientific research in the field of physical chemistry of colloids and interfaces developing theoretical models and experimental methods. He spent a decade closely collaborating with Professor Egon Matijević at Clarkson University (Potsdam, NY, USA), at first during the 1980/81 academic year, and from then until 1990, he spent three months each year at Clarkson as a visiting professor. From 1994 to 1996 Professor Kallay was the president of the Croatian Chemical Society (HKD), and since 2006 he has been editor-in-chief of the internationally renowned journal of chemical science *Croatica*

*Chemica Acta*. He has been a member of the Advisory Board of the *Journal of Colloid and Interface Science* and a member of numerous other boards and associations, such as the American Chemical Society, the Institute of Colloid and Surface Science (Potsdam, NY), the International Association of Colloid and Interface Scientists, and Institute Jožef Stefan, Ljubljana, among others.

### CROATIAN CHEMICAL SOCIETY

Professor Kallay has been a member of the Croatian Chemical Society (HKD) since 1973. With his colleague Professor Tomislav Cvitaš he began to study the problem of physical quantities and units. Within the HKD, they founded the Committee for Terminology, Units, and Symbols (KTJS), which Kallay headed from 1975 to 1979 (VII.2). In 1974 they wrote a proposal of recommendations on physical quantities and units. This proposal was based upon existing international recommendations, discussed by a large group of chemists and physicists, adopted, and published in 1975 as the official recommendation of the HKD under the title "Physical quantities and units of the international system" (*Fizičke veličine i jedinice međunarodnog sustava*). Next editions of the book were published in 1980, 1981 and 1985 (II.1). The newest edition is now being prepared for publication. The recommendations on quantities and units significantly influenced Croatian scientific literature, particularly school textbooks. N. Kallay has also worked towards legislation in the area of units of measurement, and as chairman of the Committee for Units of Measurement of the Federal Institute of measurement and refined metals (1982–84), he made sure that the text of the Law on Units of Measurement was also published in Croatian language (VII.4).

N. Kallay was a member of the Board of the Croatian Chemical Society (1976–1978, 1996–1998), as well as its president from 1994 to 1996. As president he organized a series of well-attended colloquia that were followed by get-togethers at the University Faculty Club. As an incentive for the younger generation to engage in scientific work, he established an annual award of the

HKD for young researchers aged 35 years or younger. The award was initially given for research in the field of colloid chemistry (1994–1996), and later for research in the broader field of physical chemistry (1997–2000). The sponsor of this one-thousand-dollar award was Professor Egon Matijević. This initiative prompted the establishment of yet other awards for young researchers, such as the Croatian Chemical Society's and PLIVA's Vladimir Prelog Award for organic chemistry, the award of the Croatian Biochemical Society, the Croatian state award for academic novices, *etc.* Since 2003 the HKD has presented the annual Leopold Ružička Award to the most successful young researchers in all branches of chemistry.

In 2005 Kallay was appointed editor-in-chief of the HKD's journal *Croatica Chemica Acta*. There he has strived to maintain and improve the already high level and international reputation of the journal, as well as of Croatian chemistry as a whole. He designed the logo of the Croatian Chemical Society, which was adopted and now can be seen on the cover of *Croatica Chemica Acta*.



Logo of the Croatian Chemical Society (HKD)  
Original drawing by Nikola Kallay

## THE INTERNATIONAL UNION OF PURE AND APPLIED CHEMISTRY

In 1975, at the time when the HKD's recommendations on physical quantities and units were being printed (II.1), Professor Kallay decided along with Professors Vladimir Simeon and Marijan Pribanić (Professor Cvitaš had received a fellowship to spend a year in Germany) to invite Professor Max L. McGlashan (University College, London) to come to Zagreb. In 1969, as president of the Commission on Symbols, Terminology, and Units of the International Union of Pure and Applied Chemistry (Commission I.1 IUPAC), Professor McGlashan had prepared the IUPAC *Manual of Symbols and Terminology for Physicochemical Quantities and Units* for publication. As Guggenheim's student and successor, McGlashan had dedicated particular attention to clearly defining thermodynamic concepts. The success of his visit to Zagreb is best expressed by the following excerpt from his report to the British Council:

*Among those I met in Zagreb was an astonishing young man called Dr Nikola Kallay. His understanding of rather rarefied field of algebra of physical quantities, and clarity with which he expounds it (and his lack of crankiness), are quite outstanding. As Chairman of the IUPAC Inter-Divisional Committee on Nomenclature and Symbols I regard it as most important to involve him in international work in this field and intend to do so. Such people are too rare, even in more scientifically advanced countries.*

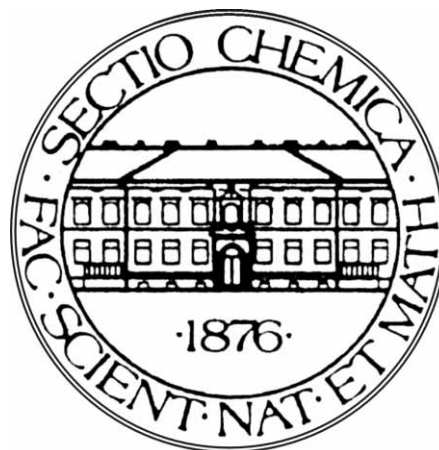
McGlashan withdrew from IUPAC, and on the basis of his recommendation, Nikola Kallay was elected titular member of Commission I.1 of IUPAC at the General Assembly of IUPAC in Warsaw in 1977. The IUPAC Manual was not sufficiently informative and thus poorly distributed, so at Davos in 1979 Kallay proposed that the IUPAC Manual undergo revision based upon the model of the HKD's Recommendations. His proposal was rejected. It was rejected once again at Leuven in 1981, but the number of opposing votes was considerably smaller. Kallay's proposal was finally adopted at the 1983 General Assembly of IUPAC in Lyngby. On that same occasion, Kallay was elected secretary of Commission I.1 of IUPAC. In addition, a working group consisting of Professors Klaus Homann, Nikola Kallay, Kozo Kuchitsu, and Ian Mills was formed for the preparation of his proposal. Soon afterwards, Tomislav Cvitaš joined the working group as well. As a result, in 1988 IUPAC published what has become its most successful publication to date: *Quantities, Units, and Symbols in Physical Chemistry*, commonly known as the *Green Book* (II.2). The manual was sold out in the same year, so a second and third printing quickly followed (1988 and 1989). Practically all well-known journals in the world use the standards of the *Green Book*, and it has been translated into seven languages. As a result of his work on IUPAC's *Green Book*, Kallay was elected to several bodies – the IUPAC Interdivisional Committee on Nomenclature and Symbols (IDCNS/IUPAC) and other related international bodies such as the International Union of Pure and Applied Physics (IUPAP), the Comité Consultatif des Unités of the Bureau International des Poids et Mesures (Sèvres, Paris), and the International Organization for Standardization (ISO) – in which he was active from 1985 until 1989, when he thanked his colleagues and withdrew from work in those bodies. He discontinued his work in the field of physical quantities and units because IUPAC's *Green Book* had already been published and he wanted to dedicate himself completely to the research in the field of physical chemistry. His work in IUPAC was continued by Tomislav Cvitaš, who as a member of Commission I.1 of IUPAC was elected secretary and eventually chairman. It was during this period that the second edition of the *Green Book* was published (1993).

## DEPARTMENT OF CHEMISTRY

In the scope of his activities at the University of Zagreb, Kallay took on various responsibilities. He began his scientific work as a research assistant at the Institute of Physical Chemistry, which later merged with the Institute of Organic Chemistry to form the Institute of Chemistry of the University of Zagreb. By governmental decision, all of the independent institutes of the University were abolished, while their employees were to be employed by divisions within the various faculties of the University, where they had been working anyway as members of the independent institutes of the University. In this complicated situation of transition, Kallay became director of the Institute of Chemistry and made sure all of the members of his institute found employment in appropriate divisions within the different faculties of the University. In 1978, Kallay himself formally joined the Laboratory of Physical Chemistry at the Faculty of Science, University of Zagreb, where he had previously worked as a member of the former Institute.

In 1982 he was elected vice-dean for finances of the Faculty of Science. This was a difficult job, because the faculty was in poor financial condition when he assumed the post. As a permanent member of the Board of the Department of Chemistry of the Faculty of Science, Kallay was devoted toward maintaining the best possible organization of work and high scientific standards. Toward this end, in 1989 he prepared the criteria for teaching and research staff promotions at the Department of Chemistry. Above other requirements, these criteria contain certain quantitative indicators that serve as minimal criteria for promotion to different academic ranks. Unfortunately, these minimal criteria later developed into a quantitative point system designating "sufficient criteria". Kallay also designed the logo of the Department of Chemistry. It is similar to the University logo, but has a picture of the oldest Chemistry building, which is located on the Strossmayer Square.

In 1991 Nikola Kallay, Vladimir Simeon, and Irenka Fischer-Palković initiated the publication of annual reports by the Department of Chemistry, which have come out each year since then. When Professor Simeon went on sabbatical in 1999, Kallay took on the duty of the head of the Laboratory of Physical Chemistry for that year. In 2001, Kallay was elected chairman of the Department of Chemistry. This coincided with all of the difficulties connected with the construction of the new Department of Chemistry building, which took years to complete. In autumn of 2005 the Department of Chemistry moved to Horvatovac. This brought all of the chemistry laboratories together under one roof, which is important for collaboration both in their common teaching responsibilities and in research. After the move, Kallay stepped down from the post of department chairman at the end of his term. Upon his appointment as chairman, Kallay had written a



Logo of the Department of Chemistry, Faculty of Science

list of principles according to which he had planned to run the department. His main tasks were as follows:

(1) to join the scattered chemical laboratories into one effective unit, thus creating a community in the area of teaching and improving conditions for cooperative scientific research.

(2) to attain as high a level of scientific research and teaching as possible through the realization of an attentive personnel policy. In spite of the high criteria in that period, seven junior associates were promoted to the rank of assistant professor.

(3) to modernize education, with the idea that the curriculum should be planned as a whole in which individual courses systematically follow one another according to a plan and are mutually interconnected. It was no simple job to put an end to the old custom of treating each course as a separate entity whose content and implementation was determined independently by each professor.

(4) to offer taught courses in specialized fields as part of the postgraduate doctoral program. When the focus of a postgraduate program is only to work on the doctoral thesis, then the result is simply the narrow specialization of the advisor, favorable only for the continuation of work on the same or similar level, rather than the all-encompassing fundamental knowledge that is necessary for true progress in scientific research and penetration into new fields.

(5) to develop collegial relations with students, for whom we should provide a program of study that is as good and as successful as possible. "University teachers exist for the benefit of the students, not the other way around."

(6) to run the department collegially and responsibly, with agreement as to the financial plan and a personnel policy that is founded upon excellence and teaching needs.



Medal of the Chemistry Department



Back of medal awarded to student Ms Nena Peran

For his results as chairman of the Department of Chemistry of the Faculty of Science, the Faculty Council awarded Kallay a certificate of recognition in April of 2006 "for his extraordinary efforts towards the development of the faculty". Nikola Kallay has dedicated particular attention to students. In order to stimulate students to work hard and to recognize outstanding achievements within the department, he had medals made that would be awarded by the Department of Chemistry at the Faculty of Science. Since 2005 the Department has awarded these medals to students in their final year who have outstanding grades. Designed by the sculptor Stjepan Divković, the medal is silver plated, with the logo of the Department of Chemistry of the Faculty of Science on the front. Engraved on the back is the name of the student and the motto "*Vivit post funera virtus*" (free translation: "Virtue does not die" or "Virtue is eternal"). The students in the Department recognized the efforts of Professor Kallay, and in response to his support and aid, in 2007 they presented him with a letter of appreciation during a meeting of the Council of the Department of Chemistry.

In this day and age, chemistry is not a very popular subject. Members of the younger generation find it difficult to decide to study chemistry. There are several reasons for this, ranging from the concept of chemistry as a profession that is responsible for the pollution of the environment (although chemistry would be capable of maintaining a clean environment) to the difficulties connected with the complexity of chemistry and the contemporary quantitative approach to the solution of chemical problems based upon mathematics and physics. For these reasons Kallay has supported chemistry presentations at University days for perspective students. In 2007 he also initiated a show entitled "Magic in Chemistry" for younger children, the intention of which was to attract them to study chemistry when they attend the university one day in the future. The show was so successful that from now

on it will be held every year both for children who have not yet encountered the natural sciences as well as for those who already study chemistry in school.

## EDUCATION IN CHEMISTRY

As a professor in the Chemistry Department, Nikola Kallay has taught, in addition to basic physical chemistry, thermodynamics, electrochemistry, and colloid and interfacial chemistry. He reorganized the student laboratories and the corresponding laboratory courses in physical chemistry and introduced an effective and collegial method of work, which has been praised by students on a number of occasions. He also introduced an advanced laboratory course in physical chemistry in which one set topic is elaborated during an entire semester, and the student has a research assistant to consult at all times. For every course that he has been involved in, Kallay has written lecture notes for his students' use. In the postgraduate program he has given lectures on kinetics and equilibrium in electrolyte solutions as well as selected chapters of colloid and interfacial chemistry. In the postgraduate programs at Maria Curie-Skłodowska University (Lublin), the University of Nova Gorica (Nova Gorica), and the Jožef Stefan Institute (Ljubljana), he has taught physical chemistry of interfaces and colloids with a particular focus on ecology.

Kallay has also promoted the teaching of chemistry in secondary schools. In connection with their work on physical quantities and units, he and Tomislav Cvitaš elaborated the quantitative approach to chemical problems based upon equations with physical quantities, instead of the concept of calculations based on proportions (and the like) that had previously been common in stoichiometry. They introduced a clear definition of the "amount of entities" and expanded it to include chemical transformations. They also pushed for the application of the concept of the "extent of reaction" and the clear comprehension of

thermodynamic quantities. His work with Professor Cvitaš in this field led to several articles published in international (VI.1,2,4,6) and national (VI.3,5) journals, but also to the publishing of several secondary school textbooks (IV.1–6). Because this new way of calculation was foreign to teachers, they held numerous seminars for them and published an accompanying booklet (IV.1) entitled "New approach to calculations in chemistry" (*Novi pristup računanju u kemiji*). Aided by the fact that Cvitaš and Kallay revised practically all school chemistry textbooks in during the 1980s, the new approach found a lasting place in Croatian schoolbooks.

## RESEARCH IN PHYSICAL CHEMISTRY OF COLLOIDS AND INTERFACES

Professor Kallay's scientific work is characterized by developments and refinements of theoretical models, the development and application of experimental methods, and the improvement of methods for the theoretical interpretation of experimental results. He does research in the field of physical chemistry, focusing on colloidal systems and interfaces. Within the physical chemistry of colloids and interfaces he has investigated the following issues:

- Electrical interfacial layer (structure, mechanism of charging, equilibrium)
- Adsorption (equilibrium)
- Aggregation of colloid particles (kinetics)
- Adhesion (kinetics, reversibility)
- Surface active substances (solubility, thermodynamics, micellization, microemulsions)
- Crystal growth (kinetics, preparation)
- Electrolyte solutions (equilibrium, ion association)
- Applied research (colloid particle preparation, drug formulation, artificial fibers, corrosion, scale deposition, ecology).

### *The Electrical Interfacial Layer*

Since the very beginning of his career as a scientist, Professor Kallay's main focus of research has been equilibrium in the electrical interfacial layer at the phase boundary. He has mainly studied solid/liquid interface, working first with silver halides and later mainly with metal oxides.

As a young scholar he ambitiously wrote three independent theoretical papers on the electrical charging of small particles (V.3–5). He introduced the "Molecular Model" and investigated colloid particles as "multivalent molecules". He solved the problem for two extreme cases, for infinitely low and infinitely high ionic strength. That restriction, with its very complex mathematics, led to the consequence that this model was never in use. The second reason was that at that time another model was

developed: the easily applicable and generally accepted "Surface Complexation Model" (Schindler 1968 and Stumm 1970), to which Kallay soon deferred. Nevertheless, it can be expected that an application of the "Molecular Model" will be found in the analysis of the distribution of charges in nano-dispersions.

Ten years after his earliest attempts, Kallay returned to the problems of electrical interfacial layers, assuming an experimental approach. He has solved the problem of the reduction of available surface (due to aggregation during experiments) by the application of ultrasound (V.37), perfected the potentiometric measurement of surface charge (III.1,V.30), and investigated the effect of the composition of liquid media (permittivity) on the charging of surfaces (V.62). Kallay has also investigated the effect of lyotropic sequence on the charging of a surface and has linked his results to coagulation phenomena (V.65,76). In their review article, Westal and Hohl (1980) clearly showed that the results from one type of experimental data (*e.g.*, surface charge density) can be interpreted with all existing variations of the original "Surface Complexation Model", and that such an approach can be used neither to confirm some concept, nor to distinguish between different concepts. Consequently, Kallay called for an as complete as possible experimental characterization of interface and the simultaneous theoretical interpretation of various types of data (III.4,5; V.124). Accepting this strategy, Assistant Professor Davor Kovačević in his doctoral dissertation (2000) elaborated the simultaneous interpretation of adsorption and electrokinetic data. He had established (III.93) that one adsorbed ion is bound to one or two surface sites, but that, due to its size, it covers (disactivates) several other sites ("umbrella effect").

Intrigued by the hypotheses of his mentor, Prof. Božo Težak (who claimed that ion pairing occurs within the interfacial layer, and that this equilibrium can be studied on the basis of Bjerrum's theory of ion pairing in a solution), Kallay formulated the equilibrium of surface association, introducing the concept of the association space being determined by the distance of closest approach and the Bjerrum critical distance. The later one is dependent on direction and on surface potential (V.42,43). Dr. Melanija Tomić solved this problem in her master's thesis, but the complex mathematical expressions were not adjusted for the interpretation of experimental results. Still, the theory answers the questions of why the association of ions is dependent upon the charge of counterions, why the association is specific (*e.g.*, in a lyotropic series  $\text{Li}^+ \dots \text{Cs}^+$ ), how the association extent depends upon surface potential, and why it diminishes in proximity to the isoelectric point.

There are several different methods for the determination of isoelectric points of surfaces, but they are ap-

plicable for materials that do not conduct electric current. When solving the practical problem of scale deposition on a heater surface, it was necessary to have a good knowledge of the isoelectric points of copper and aluminum. For these reasons, Kallay was forced to develop the so-called "Adhesion Method" for the determination of an isoelectric point by measuring the rate of adhesion of sensor particles (*e.g.*, latex) on the substrate of interest (III.7; V.34). The rate of adhesion is dependent upon the relation of sensor particle charge to the substrate (depending on pH), and the isoelectric point is determined by comparing the data for adhesion of negative and positive sensor particles. This is a well-received method that has proven useful; it has been established that the isoelectric point of metals corresponds to their oxides (V.66). This method was also developed for the monitoring of corrosion (V.79).

The potentiometric titration of a suspension leads to surface charge on a relative scale. In order to convert relative charge values into absolute values, it is necessary to establish the so-called point of zero charge (p.z.c.). The generally adopted practice rests on the assumption that the p.z.c. does not depend on the concentration of electrolytes, and it has been the custom that the p.z.c. be determined from the intersection of the surface charge function obtained at different electrolyte concentrations. This assumption only holds true if the affinity of cations and anions toward interfacial association is equal, which can be established by considering the isoelectric point. Therefore, there may be some doubt as to the validity of most published results on surface charge densities. In 1989, Noh and Schwarz developed "Mass Titration" for the determination of the point of zero charge. The method was limited to pure samples. In her master's thesis Dr. Suzana Žalac improved this method (V.70) and thereafter applied it to the investigation of the temperature dependence of p.z.c. (V.80). Mass Titration has been successfully used for mixed oxides (V.96) and has been further developed for the determination of surface charge density (V.92,122). The laboratory is currently in the process of procuring an electrokinetic instrument (acoustophoresis) that measures the isoelectric points (*i.e.p.*) of concentrated dispersions. This instrument will enable the simultaneous determination of the *i.e.p.* and the p.z.c., in order to examine the specific effects of counterions. Thus, it will be possible to test a system in order to ascertain the existence of a completely neutral electrolyte that has no influence on the *i.e.p.* or on the p.z.c. – *i.e.*, an electrolyte with equal affinities of cations and anions towards association at the interface. The aim is to find one electrolyte medium in which a certain colloid system exhibits a point of zero charge identical to the isoelectric point and the point of zero potential. Such an electrolyte may serve as a standard for a given colloid system.

Enthalpy is an important characteristic of chemical reactions. Reaction enthalpy determines the temperature dependence of equilibrium and characterizes the nature of a chemical bond. The determination of the enthalpy of surface reactions is more difficult due to the contribution of various simultaneous interfacial reactions, and also due to the electrostatic effects on those reactions. In the previous literature a few articles cite calorimetric results for the charging of interfaces (*e.g.* Machesky 1986, Lyklema 1989), but they express them as "enthalpy per charge", and not as "enthalpy per extent of specified reaction". The electrostatic effects were found to be significant. Dr. Suzana Žalac applied mass titration in her doctoral dissertation and with high precision measured the temperature dependence of the point of zero charge, which provides the standard enthalpy of surface protonation (V.75,77,80,86,107). Also, the so-called symmetrical calorimetric experiment was developed, which confirmed the results obtained by mass titration (V.75,113). The problem of the electrostatic contribution to the reaction of surface protonation, as well as that of the resolution of the contribution of the first and second stage of protonation (or protonation and deprotonation), was solved in the dissertation of Assistant Professor Tajana Preočanin on the basis of a complex interpretation of calorimetric and potentiometric measurements (III.3,6; V.94). In collaboration with Professor Wladislaw Rudzinski (UMCS, Lublin) this interpretation was improved by introducing the analysis of the surface heterogeneity (V.102,103).

The electrostatic potential of a surface – actually, the inner surface potential – directly influences the state of electrically charged species on the surface. In collaboration with Professor Miguel Blesa (CNEA, Buenos Aires), Kallay pointed out the fact that surface potential does not follow the Nernst equation, and that the slope is necessarily lower than the Nernstian slope (V.41). The significance of surface potential led to attempts at its direct measurement with the aid of metal electrodes covered with metal oxide (Penners *et. al.* 1986, and Avena *et. al.* 1993). Due to the porousness of the oxide layer, the measured potential is not necessarily surface potential, but rather the result of the redox process on the surface of the metal. Better results were obtained by the ISFET technique. On the basis of the experience of Professor Mirko Mirnik with a silver/silver iodide electrode impregnated with wax (1961), Kallay pressed hematite powder into tablets, impregnated each tablet with wax, and built it into an electrode. This experiment was unsuccessful. Then, after more than ten years, he returned to the same problem and, together with Mr. Zvonimir Dojnović, designed and constructed an ice electrode, which Dr. Duško Čakara put to use for his diploma thesis, successfully measuring surface potential at the ice/water interface (III.8; V.101). This was the first experimental result of its kind. It explained the reversible sur-

face charging of ice and, together with the well-known mechanism of fast freezing, it contributed to the explanation of the phenomenon of electrical lightning. The simultaneous interpretation of electrokinetic measurements performed in the laboratory of Professor Emil Chibowski (UMCS, Lublin) and the measurement of surface potential for ice performed in Zagreb enabled Dr. Ana Čop to explain in her master's thesis the mechanism of the charging of ice and to evaluate the corresponding equilibrium parameters (V.110,118). Using the experience with the ice electrode, Kallay then solved the problem of the porosity of metal oxides by the construction of Single Crystal Electrodes (SCrE) of hematite (V.119,123), anatase (V.124), pyrite (V.127), and other crystals. The measured surface potentials were then applied for the interpretation of interfacial equilibrium. In addition, a method for the conversion of measured electrode potential into surface potential was developed (V.125). Recently, an apparatus for measuring the surface potentials of individual crystal planes was constructed (V.129).

Kallay advocated a "General Model of Electrical Interfacial Layer" (GM-EIL) that would take into account all existing concepts and variations, but which, with the corresponding choice of parameters, could be easily reduced to the common "Double Layer" and "Triple Layer" models (III.9; V.100). In this sense he also developed a general thermodynamic approach at interface that is consistent with earlier adopted concepts related to equilibrium in solutions. He defined the standard state of interfacial species, interfacial activity coefficients and activities, and thermodynamic equilibrium constants (V.115). In terms of the latter, he showed that thermodynamic equilibrium constants are consistent with classically defined "intrinsic equilibrium constants", while he considered that the so-called "intrinsic concept" viewpoint of thermodynamics should be abandoned.

### Adsorption

In the analysis of the adsorption of organic molecules, it is necessary to establish the mechanism of their surface binding, or rather, their surface complex structure, and to ascertain the value of the adsorption equilibrium constant. Kallay approached this problem by introducing the simultaneous interpretation of electrokinetic and adsorption measurements (III.4,5; V.99,106), but later he also introduced the measurement of surface potential (V.126, 128). In addition, he analyzed adsorption equilibrium on the basis of the Surface Complexation Model. Introducing the interfacial activity coefficients and possible association or dissociation of surface species, Kallay derived the "General Adsorption Isotherm" (III.9). This general expression can in specific cases be easily reduced to the classic isotherms of both Langmuir and Freundlich. In doing so, it is not necessary to introduce restrictions

such as the non-existence of interaction between adsorbed species (Langmuir), nor low surface coverage (Freundlich).

### The Aggregation of Colloid Particles

Kallay began his work on the problem of the aggregation of colloid particles (*i.e.*, coagulation) by solving the so-called "antagonistic effect" of electrolytes (V.1,7). It was well known that a critical coagulation concentration of counterions (*e.g.*, of  $Mg^{2+}$ ) increases with the addition of electrolytes such as  $KNO_3$ , although one would expect the opposite effect – *i.e.*, the synergistic effect of  $Mg^{2+}$  and  $K^+$  ions. In his master's thesis (1972), Kallay showed that in relatively low concentrations  $K^+$  is not yet coagulatively effective but due to an increase in ionic strength the activity of  $Mg^{2+}$  ions decreases and thus increases its concentration necessary for rapid coagulation (V.1). The antagonistic effect is even more pronounced if  $K_2SO_4$  is added to the system, specifically as a result of the pairing of  $Mg^{2+}$  ions with  $SO_4^{2-}$  ions. By introducing the equilibrium constant of ion pairing and the counterion activity coefficients, Kallay solved the problem of ionic antagonism (V.7). Here it is interesting to mention that at the same time the problem of the antagonistic effect was solved by Professor Egon Matijević (Clarkson University, Potsdam, NY), who received the prestigious Kendall Award for that accomplishment.

By measuring the temperature effect on the kinetics of aggregation at high electrolyte concentrations, when the conditions for rapid coagulation are reached, Kallay confirmed Smoluchowski's theory, because the obtained apparent activation energy corresponded to the effect of temperature on the viscosity of the medium (V.6). When analyzing the effect of surface-active substances on the kinetics of aggregation, he introduced the concept of the steric contribution to activation energy (V.9). By introducing the model of surface complexation he determined the potential at the onset of the diffuse layer being responsible for colloid stability (lower than the Nernst potential but higher than the electrokinetic potential), which enabled him to successfully apply the DLVO theory (III.9; V.104). He showed that aggregation is accelerated for two reasons: because of the association of counterions with charged surface species (reducing the potential at the onset of the diffuse layer) and because of the so-called compression of the diffuse layer (III.9; V.100,104). In this way he solved the problem of lyotropic sequence in coagulation, because counterions, although they have the same charge, have a different affinity towards association in the interfacial layer (V.76).

It is well known that nano-dispersion is difficult to stabilize with electrostatic repulsive forces. Kallay approached this problem on the basis of Brønsted's solution of the primary salt effect. Since colloidal nano-par-

ticles are small in comparison to the spreading of the diffuse layer (ionic cloud), there is almost complete overlapping of their ionic clouds in the course of their collision, unlike the partial overlapping occurring with larger colloidal particles that is successfully interpreted by the DLVO theory. By introducing a transition state (of two nano-particles with a common ionic cloud), Kallay derived an expression for the stability coefficient of nano-dispersion and showed that stable nano-systems can be attained only under conditions of low ionic strength but at sufficiently high surface potentials (V.108). Therefore, it can be concluded that a stable nano-dispersion could be one which is characterized by an extremely high or extremely low isoelectric point in an aqueous medium; at  $5 > \text{pH} < 9$ , ionic strength is low  $I_c < 10^{-5} \text{ mol dm}^{-3}$ , while the surface potential is high because for example  $\text{pH}_{\text{iep}}$  is 11 or 3.

### *Adhesion*

Kallay studied adhesion mainly at Clarkson University (Potsdam, NY), collaborating with Professor Egon Matijević. He specifically studied the kinetics of the adhesion and release of colloidal particles. They solved the problem of irreversible binding of particles at surface resulting from lateral movement along an uneven surface (V.13), and explained the effects of the composition of liquid media (V.13,14), surface saturation (V.15), multi-layer deposition (V.63), *etc.* They investigated the influence of a magnetic field on adhesion and found a distinct secondary minimum dependent upon the strength of the applied magnetic field, thus enabling reversible adhesion that could be used for the separation of colloid particles (V.50,53–55). They constructed a device for examining adhesion at high temperatures (290 °C) and monitored the adhesion of small colloidal particles on relatively large spherical particles of substrate which fall freely through dispersion (V.16). As a result, Levich's theory of convective diffusion around a sphere was confirmed with a high level of accuracy. This work was recognized and nominated (as one of three candidates) for the National Association of Corrosion Engineers' Whitney Award. Adhesion experiments were also introduced at the Zagreb laboratory, where it was established that rate of release increases together with ionic strength, which points to the relaxation of interacting interfacial layers during the collision (V.38). Another method developed in Zagreb was the "Adhesion Method" for determining the isoelectric points of electrically conductive surfaces (III.7; V.34,66).

### *Surface Active Agents – Surfactants*

Kallay began to investigate surface active agents very early. That was not the main goal of his research, but over the years he would return to that problem from time

to time. First, he studied the problem of the solubility of ionic surfactants. It was established that the micellization and exchange of counterions associated with micelles determines the solubility of ionic surfactants (V.11,12,28). Together with Professor Per Stenius he applied the regular solution theory for solubility in a mixture of ionic and nonionic surfactants (V.44). Later, he studied the effect of so-called "iceberg formation" around surfactant chains in aqueous solutions. It is well known that the linear van't Hoff plot does not apply for equilibrium in surfactant systems. This phenomenon was previously explained by assuming that the extent of "iceberg formation" is dependent upon temperature. By measuring the temperature dependence of the solubility of ionic surfactants, in addition to calorimetric experiments, Kallay and Dr. Vesna Hrust demonstrated that the structuring of water around surfactant chains is not dependent upon temperature, but that the water structured around the chains has a significantly increased heat capacity (V.49,52,90). They concluded that this phenomenon is the fundamental characteristic of the surface activity. In collaboration with Professor Josip Kratochvil (Clarkson University, Potsdam, NY) the enthalpy of dilution of bile salt solutions was measured (V.36) by using the calorimeter constructed by Professor Vladimir Simeon and Mr. Mirko Tkalčec (1972).

Kallay collaborated with Dr. Vlasta Tomašić (Ruđer Bošković Institute, IRB) on the measurement of surfactant solution conductivity (V.78). Later, the activity of surfactant chains and counterions were measured by means of ion selective electrodes enabling the complete characterization of the micellization process in solutions of ionic surfactants. Introducing a molecular model of the micelle, Kallay was able to define the thermodynamic equilibrium constant for micellization and to determine its value, as well as for the association of counterions with charged micelle chains (V.132). Kallay collaborated with Dr. Alba Chittofrati (Ausimont, Milano) on the problems of microemulsions, and refined the fluctuation theory developed by Eicke and Borkovec (1989) for the conductivity of microemulsions (actually, nanoemulsions), by distinguishing the radius of the internal drop of water from the entire radius that includes the surfactant chains connected to a nano-droplet of water (V.59). Later, Kallay worked together with Dr. M. Tomić on the permittivity effect of nano-droplets of water, for which it was necessary to solve a concave model of a spherical electrical interfacial layer (V.71).

### *Crystal Growth*

The kinetics of crystal growth and the preparation of colloidal particles are problems that Kallay has dealt with on the side. In 1983 Dr. Darko Babić and Mr. Zvonimir Dojnović constructed an on-line computer-con-



trolled potentiometric titrator that made it possible to monitor the kinetic crystallization of oxytetracycline (V.24). Later, Kallay collaborated with Dr. Ljerka Brečević (IRB) on the kinetics of the dissolution of calcium carbonate, which they monitored calorimetrically (V.87). With Ms. Irenka Fischer, M. Sc., he introduced a continuous flow method for the preparation of monodisperse spherical particles of hematite (V.23). A laboratory HEPRO apparatus was constructed on the basis of which E. Matijević at Clarkson University developed an apparatus of greater dimensions that was successfully put to use and patented.

### *Electrolyte Solutions*

In his diploma thesis, Kallay dealt with the complexation of  $\text{Cd}^{2+}$  ions with EDTA. In his master's thesis, he solved the problem of the so-called "antagonism" of ions in coagulation processes, considering the association of ions in solution and elaborated a method for determining the equilibrium constant of ion pairing by means of coagulation measurement (V.7). In this way, he determined the stability constants of the ionic pairs  $\text{LaSO}_4^+$  and  $\text{MgSO}_4$ . Later, he assisted in the research of Professor Vladimir Simeon in the investigation of ion hydration in aqueous solutions (V.111). With Assistant Professor Vladislav Tomišić he was involved in investigation of the enthalpy of ion association on the basis of temperature dependence of the equilibrium constant and calorimetry.

### *Applied Research*

Kallay has applied his fundamental research towards finding solutions to practical problems in technology and elsewhere. His motivation is twofold. He finds it interesting to apply the results of fundamental research in order to solve practical problems. In addition, however, the funds received from applied research projects enable the laboratory to acquire better equipment. Together with Professor Mira Bećirević (Faculty of Pharmacy, Zagreb), Kallay applied his experience on kinetics of crystal growth and dissolution to the formulation of medicines with prolonged effects (V.21,45). He also has collaborated with Professor Ana Marija Grancarić (Faculty of Textile Technology, Zagreb) on solving a problem related to the kinetics of the dissolution of synthetic fibers, which is important in the technology of artificial silk (V.61,72). Kallay has dealt with the problem of scale deposition in heaters, finding conditions in which the deposition is significantly reduced. An added result of this investigation was the development of a method for the determination of the isoelectric point of metals (V.34). Kallay adapted this method for the monitoring of the process of corrosion (V.79). With Dr. Jasenka Bišćan (IRB), he investigated the electrokinetic properties of silicon nitride in non-aqueous solutions, which is impor-

tant in the technology of synthetic ceramics (V.86,98). In cooperation with Forschungszentrum Jülich, he dealt with ecological problems by studying the adsorption/desorption equilibrium of model organic pollutants in the soil.

### FUTURE PERSPECTIVES

Professor Kallay has always led a relatively small research team because his policy was to be involved in all details of the research. However, as can be seen from the list of publications, he has had extensive collaborations with numerous research groups in the world. Most recently, his team has developed two aspects of colloid and interfacial chemistry. Assistant Professor Davor Kovačević, after returning from his postdoctoral studies in Wageningen, where he collaborated with Professor Martien Cohen Stuart, has moved to the field of macromolecules and biocolloids. He has organized his laboratory in Zagreb and has already published several articles which have been recognized by the international scientific community. Assistant Professor Tajana Preočanin remained in her original area of inorganic systems, and is involved in the development of methods for investigating interfacial equilibrium. In her work, she is collaborating with Dr. Johannes Lützenkirchen from Forschungszentrum Karlsruhe applying our recently developed method for the measurement of the surface potentials of the individual crystal planes.

During his career, professor Kallay aimed at solving some fundamental problems but also at applying fundamental knowledge in solving practical problems. His work is respected, and his articles are well cited. Most recently, Kallay has been involved in a unique international project: His research on the electrical interfacial layer and his analysis of a variety of existing concepts and controversies – including the mechanism of interfacial reactions, the definition of the interfacial standard states and equilibrium constants, the concept of capacitors of constant capacitances, and the concept of electrokinetic slip plane – led him in 2007 to organize a Discussion Meeting in Opatija, on the northern Croatian coast, in cooperation with Dr. Johannes Lützenkirchen (Forschungszentrum Karlsruhe) and Dr. Vinzenz Brendler (Forschungszentrum Dresden-Rossendorf). This meeting was organized so that each entire session would be dedicated to an individual problem, and that in each discussion the goal would be to seek a "common ground" and to define the points of controversy. The meeting was appreciated and fruitful. One of the goals was to agree on the practical approach in the interpretation of interfacial equilibrium. Further scientific development in the field is expected from the development of theoretical models, as well as from more advanced and refined experimental methods (e.g., spectroscopy). However, from

the practical point of view, one needs to decide on the common approximate model in order to interpret the data so that the equilibrium parameters are comparable. A compromise (Opatija Convention) was reached by assuming two independent 1-pK mechanisms ("Double 1-pK Model"), thermodynamic equilibrium constants based on interfacial activities (corrected for electrostatic interactions), constant capacitance of the inner Helmholtz layer of  $1 \text{ F/m}^2$ , and the total concentration of active surface sites of  $10^{-5} \text{ mol m}^{-2}$ . With respect to the diffuse layer, the Gouy Chapman theory will be used together with the bulk permittivity of solvent. The incorporation of the electrokinetic data has two options. One is to apply the Triple layer Model (TLM) and to assume a second layer of constant capacitance, and the potential at the onset of diffuse layer equal to the electrokinetic potential. The second approach is based on the Double Layer Model (DLM), which neglects the second layer but introduces the distance of the electrokinetic slip plane separation of 1 nm. The results of the discussions are to be published in a special issue of the journal *Croatica Chemica Acta*.

## AWARDS & RECOGNITIONS

Professor Nikola Kallay, whose work is recognized by the scientific community and his students alike, has received the following awards.

1. The Ruđer Bošković Award of the Republic of Croatia for scientific achievement, 1992.
2. The Award for Scientific Achievement of the Croatian Academy of Sciences and Arts, 1999.
3. The Award of the City of Zagreb for scientific accomplishments, 2000.
4. Elected associate member of the Croatian Academy of Sciences and Arts, 2006.
5. Certificate of Recognition presented by the Council of the Faculty of Science, University of Zagreb, 2006.
6. Letter of appreciation from the students of the Faculty of Science, University of Zagreb, for his help and support, 2007.

*Davor Kovačević  
Tajana Preočanin*

## Nikola Kallay – List of Publications

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