Chronicle

Vytautas Kaikaris
(May 22, 1912 – August 29, 1982)

Ninety years have passed since the birthday of Vytautas Kaikaris, the famous Professor of Vilnius University, and twenty years will have passed this year since his death.

Vytautas Kaikaris was born on May 22, 1912 to a family of a pharmacist in Kovarskas, Anykščiai district. He was the eldest of three brothers who all became scientists – two chemists and one pharmaceutist-chemist. V. Kaikaris finished the gymnasium of Žagarė in 1927, the teacher-training seminary of Šiauliai in 1931, graduated from the Physics Cycle at the Faculty of Mathematics-Nature of the Vytautas Magnus University (Kaunas) in 1936 and was conferred a diploma of a physicochemist. In autumn 1936 he was invited to work at the Department of Physical Chemistry and Electrochemistry of the Vytautas Magnus University as a junior laboratory assistant, in 1937 became a senior laboratory assistant of the same Department and in 1939 – a junior assistant.

Under the scientific supervision of the Head of Department J. Matulis, he began investigations on the kinetics of Menschutkin reactions (the combination of alkilhalogenide with tertiary amine in different solvents). When Lithuania recovered its capital Vilnius in 1940, V. Kaikaris together with the Faculty of Mathematics-Nature moved to the Vilnius University, where he fulfilled the duties of the junior assistant at the Department of Physical Chemistry for two years. After maintaining a doctoral thesis "The kinetics of the combination reaction of ethyliodide with triethylamine" in 1942, he was appointed a senior assistant. When the German occupation authorities closed down all higher schools in Lithuania on March 12, 1943, V. Kaikaris withdrew to the provinces and in 1943–44 worked as an assistant of the chemist and brewer in the Mažeikiai Brewery.

After the end of the Second World War, V. Kaikaris returned to Vilnius again and in 1944 was appointed Head of the Department of General Chemistry at the Vilnius State University. He fulfilled these duties till 1956, delivered lectures on physical and colloid chemistry, general and inorganic chemistry, physicochemical analysis and even on organic chemistry for the students of Medical Faculty, Faculty of Natural Sciences and others.

Unfortunately, Soviet authorities did not recognize a doctoral thesis maintained under German occupation, so Vytautas Kaikaris had to defend his thesis for the second time in 1947; a candidate of science (now doctor) degree was conferred on him, the first among the chemists of the Vilnius University.

In 1946–1950 V. Kaikaris was the Dean of the Faculty of Chemistry. In 1953 the academic degree of Docent (Assistant Professor) was conferred upon him. From 1956 to his death in 1982 (*i.e.* for 26 years) V. Kaikaris was the Head of Department of Physical Chemistry, delivered the main course of lectures on physical chemistry for chemists, supervised diploma works of students and dissertations of assistants and post-graduate students.

At the beginning of his self-dependent researches, taking into consideration the dramatically grave postwar situation and a poor supply of the University with more or less complicated equipment, V. Kaikaris undertook to solve some problems of physico-

chemical analysis. There were just the theoretical fundamentals and practical possibilities of physicochemical analysis investigated by the first two of his disciples – Kostas Januševičius and Algimantas Levinskas.

K. Januševičius in his thesis "Differential potentiometric determination of halides, rhodanide and calcium ions" (in Lithuanian) maintained under the supervision of V. Kaikaris in 1958 studied the possibility to employ a mercury indicator electrode for determination of halides and rhodanide as well as their various mixtures by a titration with solutions of mercury nitrate. An indicator electrode of the third kind $Hg|Hg_2C_2O_4$, CaC_2O_4, Ca^{2+} likewise was investigated in an effort to determine calcium in natural substances such as limestone, dolomite, etc. The analytical feasibility of the method, the limits of experimental error, the blocking ions were defined.

A. Levinskas in his thesis "Potentiometric determination of calcium and magnesium using electrodes of the third kind and a bismuth electrode" (in Lithuanian) maintained under the supervision of V. Kaikaris in 1959, examined in detail the electrodes of the third kind of the simplest construction ($Pb(Hg)|PbC_2O_4$, CaC_2O_4, Ca^{2+} and $Hg|HgC_2O_4$, CaC_2O_4, Ca^{2+}) in solutions of calcium chloride and nitrate. He disclosed their advantage over the membrane electrodes of the third kind and the possibilities of their employment in chemical analysis. The potentiometric method was prepared and tested in practice for determination of calcium and magnesium without preliminary separation of these ions, but using two indicator electrodes: the third kind and a bismuth one.

When in the sixth decade of the 20th c. the Institute of Chemistry and Chemical Technology of the Lithuanian Academy of Sciences under the guidance of Prof. J. Matulis undertook to concentrate researches in the field of theoretical and applied electrochemistry and all scientific investigations had been subjected to co-ordination, V. Kaikaris turned his researches also to electrochemistry, predominantly to the problems of silver electrodeposition.

The first investigations in the field of electrochemistry performed under his guidance were summarized in the thesis of Anatolij Voronko "Studies of surface levelling at metal electrodeposition" (in Russian) defended in 1963. In this work, microdistribution of nickel, copper and zink on the surface during electrodeposition of these metals from electrolytes of various composition without levelling agents was investigated. The geometrical levelling of electrodeposits was found to be improved by the factors that decrease the concentration polarization: a decrease of current density, a rise of temperature, an increase of concentration of discharging ions, etc. The levelling agents may change customary levelling regularities. The effect of each factor on the levelling capacity depends upon how a change of this factor is reflected on the run of the "current density – concentration of additive" curve at a constant potential.

In 1964 Algirdas Kundra defended the thesis "An investigation of complex chloride and bromide solutions of silver plating" (in Lithuanian). That was the first dissertation maintained under the guidance of V. Kaikaris in the field of silver electrodeposition, to which V. Kaikaris devoted all his subsequent researches: all his other post-graduate students maintained their theses concerning solely the problems of silver electrodeposition; thus V. Kaikaris found his own niche in the electrochemical studies extended greatly at that time. The above-mentioned thesis of A. Kundra examined the possibilities of complex chloride and bromide electrolytes of silver plating in obtaining silver electrodeposits of various character, and the optimum composition of electrolyte and operating conditions were recommended. Relying on abundant evidence, an original view on the mechanism of silver electrodeposition from complex bromide solutions was proposed.

Irena Pivoriūnaitė, a post-graduate student of V. Kakaris, in her thesis "Silver electrodeposition from solutions of potassium dicyanoargentate" (in Lithuanian) defended in 1965 examined thoroughly the electrode processes in potassium dicyanoargentate solutions without "free" cyanide in an effort to obtain less toxic electrolytes. In parallel with the theoretical investigation of various electrode processes, a cyanide-rhodanide electrolyte without free cyanide was proposed to obtain bright silver coatings, which was tested under industrial conditions.

The search for less toxic electrolytes was continued in the thesis of Sigita Pilauskienė, another post-graduate student of V. Kaikaris, "Study of silver electrodeposition from pyrophosphate electrolytes" (in Lithuanian) which was defended in the same year 1965. The author investigated the influence of various factors on the cathodic polarization and proposed a plausible scheme of the mechanism of silver electrodeposition from pyrophosphate solutions. It was found that under certain conditions mirror-bright silver coatings might be deposited from especially pure phosphate solution. Furthermore, a pyrophosphate solution for silver electropolishing was proposed.

Continuing this line of research under the guidance of V. Kaikaris, in 1968 the thesis of Teofilis Jankauskas "An investigation of dicyanoargentate–rhodanide electrolyte" was maintained, in which the peculiarities of cathodic processes and the influence

of various factors upon them, the influence of various special additives on the physicochemical properties of electrolyte as well as on the course of electrode reactions and on the properties of the silver electrodeposits obtained were examined. All the regularities established in the dicyanoargentate-rhodanide electrolyte were compared to these of cyanide solutions considered then as more superior.

Further rather close investigations of this line were maintained in 1970 in the thesis of Vytautas Skučas "Studies of silver electrocrystallization in complex electrolytes" (in Lithuanian). In the course of this investigation, a new type of silver electrolyte containing two ligands (cyanide and rhodanide ions) which could form various silver complexes was elaborated. With the aid of a substantial variety of electrochemical methods, three potential regions of different cathodic processes were distinguished and the most credible schemes of silver electrodeposition mechanism in different potential regions were proposed. An interrelation between microspheroid coatings and the formation of cathodic films containing carbon was established. The silver rhodanide-cyanide electrolyte was demonstrated to be a convenient model for the examination of the mechanism of metal electrocrystallization when various complexes of one metal are present in a solution.

In 1969 V. Kaikaris summarized the numerous investigations performed in collaboration with his disciples and maintained the thesis for the then degree of doctor of sciences "Studies of silver electrodeposition processes". In this work he systematized data on silver electrolytes of six types distinguishing considerably by the prevailing silver ions of different composition: (1) solutions of silver nitrate containing hydrated simple silver cations, (2) solutions of silver sulphamate containing silver hydrosulphamate anions, (3) complex pyrophosphate solutions containing hydrated diaminosilver anions, (4) solutions of silver lactate in which part of silver is in the form of an undissociated salt and part is in the form of hydrated Ag^+ cations, (5) solutions of silver halogenides and silver rhodanide in which silver exists in the form of hydrated complex anions $AgA_n^{(n-1)-}$ where A is a corresponding halogenide or rhodanide anion and n = 2, 3 or 4, (6) various cyanide elecrolytes with a insignificant content of "free" potassium or sodium cyanide in which dicyanoargentate ions prevail.

From the investigation of these electrolytes V. Kaikaris drew the conclusion that silver electrocrystallization may proceed in three different ways: 1) according to the classical electrocrystallization mechanism, *i.e.* reduction undergo either (a) positive ions (*e. g.*, Ag^+, or (b) neutral molecules (*e. g.*, AgBr, AgCN, *etc.*) or (c) negative ions (*e. g.*, $Ag(CN)_2^-$); 2) reduction undergo suspended or colloid particles, *e. g.*, $(AlCl)_s$, $(AgCN)_s$, $(Ag_2S)_s$, *etc.*; 3) reduction undergoes the gelatinous film as in the sulphamate, pyrophosphate or lactate silver electrolytes.

In his dissertation V. Kaikaris paid much attention to the structure and surface morphology of silver electrodeposits. Developing the classification of cathode deposits elaborated by J. Billiter and H. Fischer, he proposed a new classification of electrodeposits evolved from his own optical methods devised from the examination of metal surface morphology. The main criterion of this classification is assumed to be the degree of crystallinity and dispersity of electrodeposits. On this principle, all electrodeposits may be divided into three large groups: 1) clearly crystalline, 2) UD type (according to H. Fischer) and 3) cryptocrystalline deposits. Deposits of the first group were additionally subdivided into: a) dendrites, b) isolated crystalls, c) coarse-crystalline deposits and d) fine-grained deposits. Each of these three deposit groups is related, according to V. Kaikaris, to the three above-mentioned crystallization ways.

Another important and original idea examined in this work was the so-called two-factors theory of the formation of bright coatings. Its basic sketch was reported at the conferences on electrochemistry in Riga (1965), Vilnius (1966), Dnepropetrovsk (1967), and an extended discussion was published in the journal "Elektrokhimiya" in 1967. In V. Kaikaris' opinion, the effect of two factors is required for the formation of bright metal coatings. These are: 1) hindrance of crystallization process due to inhibitors adsorbed on the crystallization centers and hindering their subsequent growing, or due to a precipitation of colloid particles on the cathode, and 2) formation on the cathode surface of a gelatinous film which can be reduced by the electrons or by adsorbed hydrogen. On the basic of this theory, a new classification of brighteners was also proposed, dividing all brightening agents into two large groups. To the first group there were attributed compounds that cause a formation of colloid particles capable of reduction on the cathode, and to the second group comprised compounds, mainly macromolecular, that have the capability of the formation of phase films of optimum thickness and viscosity on the cathode surface. Admittedly, his former teacher Acad. J. Matulis was rather critical about this theory, although it was accepted by some Russian scientists (*e. g.*, I. D. Kudriavtseva, I. N. Andreev).

Thanks to the mentioned works, V. Kaikaris became a recognized authority in the field of silver electrodeposition problems not only in Lithuania, but also beyond it – in the other republics of the former Soviet Union. In 1970 the title of Professor was conferred upon him.

Once having called attention to the formation of colloid particles and of cathode film in the course of silver electrodeposition, V. Kaikaris also after defending his doctoral thesis made an effort to explain the effect of such a film on the cathodic polarization as well on the structure of electrodeposited metal. In 1972, by his consultation and under the supervision of Doc. J. Januševičienė, the post-graduate student Leonas Adolfas Sakalauskas maintained his thesis "Studies of cathodic processes in solutions of copper and silver salts" (in Lithuanian), in which attempt was made to simulate the electrode processes employing an artificial film of agar gel on the cathode in solutions of various copper and silver salts.

When examining the processes of silver electrodeposition, V. Kaikaris became naturally interested in the anodic processes, too. Together with his former disciple Doc. T. Jankauskas he supervised the thesis of Petras Juzikis "Studies of silver anodic processes" (in Russian) maintained in 1977. In this work the effect of many factors on the kinetics of silver anodic dissolution was considered in various electrolytes, and the nature of the limiting currents was established. The composition of the prevailing complexes in the dicyanoargentate-rhodanate electrolyte was examined and the mechanism of the anodic process was proposed. Also, the formation conditions of passivating films on the silver anode and the chemical composition of these films were determined. The behavior of the so-called "composite" electrodes (silver–graphite, silver–antimony, etc.) was studied in dicyanoargentate-cyanide silver electrolyte, and it was proposed to utilize these electrodes for adjusting the electrolyte composition. A new silver electropolishing process was developed, using anodic impulses of high current density.

Alongside the electrodeposition of pure silver, Prof. V. Kaikaris was also interested in the electrodeposition of silver alloys. Together with Doc. T. Jankauskas he supervised the thesis of Vytautas Daujotis "An investigation of electrodeposition of some silver alloys" (in Russian) maintained in 1980. In the course of this work the effect of organic sulfur surfactants on silver electrodeposition from complex cyanide solutions was established and investigated. The established regularities were applied for regulation of silver deposition potential in an attempt to deposit silver alloys with other metals. For investigation of the inhibiting adsorption of the surface-active substances studied on the silver electrode, a new ellipsometric method elaborated by the authors themselves was employed. Using this method, the thickness of adsorption films forming on the silver electrode was determined. The results of this investigation enabled elaboration of original electrolytes to obtain coatings of silver–antimony and silver–nickel alloys.

Finally, with one of his disciples, Doc. V. Skučas, Prof. V. Kaikaris supervised a thesis of Pranciškus Varkala "An investigation of electrodeposition of silver and its alloys with indium and thallium from the dicyanoargentate-rhodanide electrolyte" (in Russian) which was maintained in 1983, already after the death of Professor. In this work the kinetics of electrodeposition of silver–indium and silver–thallium alloys from the dicyanoargentate-rhodanide electrolyte was examined. The conclusion was made that while electrodepositing a silver–indium alloy the products of partial reduction of indium ions are generated at the cathode. The formation of a silver–thallium plating alloy is accompanied by a pronounced cathodic depolarization of the process, which may be caused by chemical cementation of silver by thallium and in addition by activation of the cathode surface. An electrolyte was elaborated for deposition of hard, anticorrosive, friction-resistant, bright silver–thallium coatings. An higher microhardness of silver–thallium coatings (in comparison to pure silver) was supposed by the authors to be due to inclusion of nonmetallic particles into the electrodeposits. The complex compound $Tl_2Ag(CN)_3$, earlier unknown, was discovered and its synthesis and analysis were described. The silver preplating noncyanide electrolyte elaborated by the authors and designed for the surface superfinishing of bronze and other copper alloys, metals of iron subfamily and their alloys, tin and its alloys and some other metals before silver or gold electrodeposition without the copper underlayer was introduced into production. Also, an original method of silver regeneration was elaborated, which is characterized by universality, by totality of silver separation from solutions of its cyanide complexes and by the advantages that highly toxic hydrogen cyanide is not evolved in the course of regeneration: the free cyanide is bound and the dicyanoargentate complex is destroyed by the aldehyde, whereas silver is precipitated in the form of a slightly soluble compound.

Prof. V. Kaikaris may be considered a founder and a leader of the independent electrochemists' school of the Vilnius University. His scientific researches were conducted in the united field of investigation of silver electrochemical processes. Under the scientific supervision of V. Kaikaris alone or with his co-workers 12 theses were maintained for the former candidate's (now doctor's) degree. He published, together with his co-workers, 114 papers, reports and other scientific works, received 13 patent certifications of the USSR. He made reports at numerous scientific conferences in Lithuania and in

other republics of the former Soviet Union. Two of his disciples (A. Levinskas and V. Daujotis) themselves maintained theses for a habilitated doctor's degree, became professors, founded their own scientific schools, to tell the truth, moving somewhat away from the research problem of their teacher, but remaining devoted electrochemists. It is a pity that the scientific works of Prof. V. Kaikaris, which were published in the beginning mainly in Lithuanian and later in Russian, because of the political situation at that time were little known in Western countries and consequently did not receive a proper attention and comment.

Prof. V. Kaikaris was a gifted teacher, a well-informed lecturer. He delivered the main course of lectures on physical chemistry for more than two decades at the Department of Physical Chemistry of the Vilnius University. His lectures were comprehensible, clear, not overcharged with unnecessary details and with useless over- theorized discourses.

As the Head of Department, Prof. V. Kaikaris was rather democratic, did not thrust his opinion on junior colleagues, but tried always to convince them of the correctness of his ideas. He often visited the lectures of his co-workers, not infrequently without notifying them beforehand, however, he set forth his remarks and advises after visitation tactfully tête-à-tête. Thus he was able to create a calm and encouraging climate in the department for carrying out both the pedagogic work and scientific researches. Professor often perceived even low spirits of his colleagues and did his best to help in grave situations of their life or work. As a well-known expert in his research field, he was appointed an official opponent to about 60 theses including several doctorals degrees (habilitation). In his evaluation of scientific works, Prof. V. Kaikaris was objective, emphasized both the positive and the weak points of a dissertation, but he was always benevolent, avoiding undesirable discussions on the questions of minor importance and appreciating the innovation of the work and the findings of new common regularities. On the other hand, being very accurate, careful and sometimes even pedantically orderly, he claimed the same from others.

Prof. V. Kaikaris by his actions and works won a profound respect of many of his disciples, colleagues and acquaintances, left warm reminiscences in our memory.

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Prof. L. Simanavičius

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Patents

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