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The Importance of V.G. Levich's Research in the Development of Modern Electrochemistry[1, 2]

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Abstract—The fundamental scientific areas founded and developed by V.G. Levich and his school, and their importance in contemporary theoretical electrochemistry have been overviewed.

Keywords: physico-chemical hydrodynamics/mechanics, quantum mechanical theory of charge transfer, mass transport, bioelectrochemistry

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Veniamin Grigorievich Levich's primary legacy is that he created a whole new field of science at the border between physics and chemistry, "Physico-Chemical Hydrodynamics/Mechanics." The former term was used by Levich himself as the title of his famous monograph, first published in Russian in 1952 and (as a second edition) in 1959, then translated into English and published in 1962 [1]. Veniamin G. Levich was, however, a researcher of prodigious originality and productivity in a wider impressive breadth of areas, more adequately described as "Physico-Chemical Mechanics" that ranged from electrochemistry to turbulence, transport in porous media, to bubbles and particles, capillary phenomena, photoemission of electrons from electrodes into electrolyte solution, and last, but not least the first quantum mechanical theory of electron transfer between molecules in solution and between molecules and electrochemical electrodes [2–19].

The scientific achievements of V.G. Levich and his school have had a profound impact in many areas of modern theoretical electrochemistry. In particular, these areas include: Quantum theory of the elementary act of electrochemical processes; theory of electrical currents through electrolyte solutions; bioelectrochemistry; theory of mass transport across diffusion layers and of electrochemical noise under conditions of the turbulent flow; theory of the diffusion layers with a spatially-extended electrical charge; theory of ion transfer processes in electrochemical systems with particular reference to the rotating disc and rotating ring-disc electrodes and to the extending diffusion layer along planar electrode surfaces; theory of "the dynamic psi-prime effect" (for the electrochemical reduction of anions); theoretical problems in the physics and chemistry of dispersed systems in moving liquids and gases; theory of the motion of drops, bubbles and solid particles in liquid media and electrolyte solutions; theoretical problems in flow and diffusion in thin liquid films; structure of electrified metal/solution interfaces; specific ionic adsorption; theory of the operation of electrochemical transducers of mechanical characteristics into electric signals; and, theory of the noise of electrochemical systems, electrochemical noise diagnostics, and noise-registration based methods in search for oil and gas fields [20–34].

Veniamin G. Levich was born in Kharkov in 1917, a turbulent period of Russian history. At the age of twenty he acquired his first university degree from Kharkov University. At this university he met the great theoretical physicist and Nobel laureate to become,

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[2] The article is published in the original.

Lev D. Landau. He then enrolled as a Ph.D. course at the V.I. Lenin State Pedagogical Institute in Moscow with Landau as his supervisor. Those of us who had the privilege of knowing V.G. Levich closely remember him telling about the "Landau's theoretical minimum" entrance exam, which each young candidate who dreamt of becoming a Ph.D. student of Landau had to pass. Landau gave him the task of calculating a tricky integral which required transformation with the use of the complex variable plane. The applicant would then have to describe all steps of this solution orally in detail to Landau. Paper, writing utensils, or blackboard were not allowed, and the examination session was running whilst Landau and the candidate would walk together along the corridor. V.G. Levich passed the test successfully in 1937 to enroll as a Ph.D. student of Landau. His Ph.D. thesis, "On the theory of surface processes," was defended in 1940. Three years later V.G. Levich defended his Doctor of Science Dissertation (equivalent to the Habilitation Degree in Europe) "On current flow through electrolyte solutions." These studies already then led him to the importance of concentration polarization and triggered his later development of physical–chemical hydrodynamics and the renowned research tool, the rotating disc electrode.

V.G. Levich's especially productive years began, when shortly after his Ph.D. defense he joined Academician A.N. Frumkin at the Colloidal-Electrochemical Institute (renamed Institute of Physical Chemistry in 1945), and then at the Institute of Electrochemistry (renamed later into the A.N. Frumkin Institute of Electrochemistry, and still later the A.N. Frumkin Institute of Physical Chemistry and Electrochemistry), all belonging to the Academy of Sciences of the USSR or to the Russian Academy of Sciences. He was the creator and head of this institute's theoretical department in the period of 1958–1972 and also a full professor and head of department at the Moscow Institute of Engineering Physics (MIFI) 1954–1964, as well as full professor and head of the Chemical Mechanics chair at Lomonosov Moscow State University (MGU) 1964–1972. In 1978 he accepted prestigious full professorships first at the University of Tel Aviv in Israel and the following year at the City University of New York where he created and became director of a new institute, The Institute of Applied Chemical Physics today known as the "Veniamin Levich Institute of Physico-Chemical Hydrodynamics."

During the period of his stay at The Institute of Electrochemistry of the Academy of Sciences of the USSR, 1958–1972, Veniamin Grigorievich Levich was not only an outstanding scientist but also showed excellent talents in scientific organization and management. As head of the theoretical department of this institute he gathered an extremely strong group of theoretical physicists and physical chemists in multifarious areas of physico-chemical mechanics and theoretical physics of electrochemical systems. Of immense importance for the research of the theoretical department were the famous "theoretical seminars," which attracted dozens of participants from the institute itself and from other academic and industrial institutions to this vibrant environment of new theoretical work. The seminars took place weekly and were always accompanied by extremely hot and exciting critical discussions.

In 1958 an extraordinarily talented young Georgian theoretical physicist, Revaz Romanovich Dogonadze came to V.G. Levich's group after his graduation from MIFI. Revaz Dogonadze had begun his work from theories of electron transport in structurally disordered systems and of electronic relaxation in solids. This area had grown out of solid state and semiconductor physics, which had emerged from Second World War efforts, with Kubo in Japan, Lax and Holstein in the USA, and Pekar in Ukraine as prominent figures. Experimental studies of simple, outer sphere chemical electron transfer processes in solution, say between transition metal complexes, had begun at about the same time with Henry Taube in the USA as a pioneering figure.

Levich and Dogonadze were the first [35] to view condensed matter chemical electron transfer processes in a way similar to electronic relaxation processes of localized impurity states in polar solids, i.e. as a quantum mechanical transition between two separate, weakly interacting electronic states each strongly coupled to a vibrational continuum. The immediately appealing renowned equation for the "non-adiabatic" ("diabatic") electron transfer rate constant k (s^{-1})

$$k = \frac{2\pi}{\hbar}\frac{(V_{fi})^2}{\sqrt{4\pi\lambda k_B T}}\exp\left(-\frac{\Delta G^{\neq}}{k_B T}\right), \tag{1}$$

was the primary result already in their 1959 paper, shortly after generalized to the adiabatic limit and intermediate regimes in equally appealing transparent form

$$k = \kappa\frac{\omega}{2\pi}\exp\left(-\frac{\Delta G^{\neq}}{k_B T}\right), \tag{2}$$
$$\kappa_{\mathrm{diab}} \ll 1;\quad \kappa_{\mathrm{intermed}} < 1;\quad \kappa_{\mathrm{adia}} \to 1.$$

An amazing foresight was included already in their 1959 pioneering paper [35] which addressed for example the notion of potential energy surfaces, low-temperature processes, and nuclear tunneling. Following these early pioneering joint studies, Levich established a new group headed by Dogonadze in the theoretical department of the Institute of Electrochemistry. This led to a more than twenty years' period of intense and enthusiastic efforts, in which an impressive range of chemical and electrochemical processes were given radically new theoretical frames.

These included for example, extension to electrochemical "elementary" processes at metal and semiconductor electrode surfaces, induced by environmental polarization fluctuations, and extension to electron transfer processes accompanied by intramolecular nuclear reorganization [36, 37]. A radically novel view of proton transfer reactions between a donor and acceptor molecular fragments was also proposed. The decisive role of the proton tunneling phenomenon rooted in the much larger proton vibrational energy quanta compared with the thermal energy [38] was much more prominently recognized in this paper than earlier. This view was at first met with open skepticism but gradually became the generally accepted view of chemical and electrochemical proton and hydrogen atom transfer theory and was in fact later rediscovered and given very similar forms by scientists elsewhere. Other efforts led to formal theoretical correlations between (electro)chemical electron transfer and other molecular electronic processes such as optical and even biological processes. Key figures in Dogonadze's team in this development in the late 1960's and the 1970's were Yurij A. Chizmadzhev, Alexander M. Kuznetsov, Mikhail A. Vorotyntsev, Alexei A. Kornyshev, Erik D. German, and Yurij I. Kharkats, who all became internationally recognized scientists. Foreign scientists such as Jens Ulstrup, Wolfgang Schmickler, and Ronald W. Fawcett also spent extended research stays at the theoretical department of the Frumkin Institute in this "heroic" period.

In retrospect, the pioneering contributions of Levich, Dogonadze, Kuznetsov, and their associates to chemical and electrochemical molecular charge transfer theory were unfortunately hampered in the early pioneering days by the imposed need for publication in journals which were at the time not easily accessible outside the USSR. Strict space limitations in the leading Soviet journals was also an obstacle. Many of these results have therefore been wrongly credited to much later work elsewhere which will hopefully be fully recognized.

V.G. Levich's work on physicochemical hydrodynamics and quantum mechanical molecular charge transfer theory have left a lasting impact. His renowned monograph, "Physico-Chemical Hydrodynamics," described most broadly the effects of fluid motion on physical chemical processes and vice-versa. The book is a masterful synthesis of different areas of physics and physical chemistry accompanied by elegant mathematical frames and till the present day a delight to read, now a true classic.

V.G. Levich was in fact an excellent book writer. Inspired by his great mentor L.D. Landau, he wrote the two-volume Treatise of Theoretical Physics as a shorter and more easily readable book for students and young researchers, compared to the fundamental multi-volume series by L.D. Landau and E.M. Lifshits. This masterpiece of V.G. Levich has retained its usefulness till now, and is delightful reading to study chapter by chapter.

V.G. Levich received national and international, rightfully deserved recognition. In addition to his corresponding membership of the Academy of Sciences of the USSR he was awarded the Palladium Medal of the American Electrochemical Society in 1973. He was elected a foreign member of the Norwegian Academy of Sciences in 1977 and an Associate of the US National Academy of Engineering in 1982. He passed away in 1987, 70 years of age. By his impressive scientific achievements in a range of areas spanning electrochemistry, hydrodynamics and mass transport, and quantum mechanical charge transfer theory, and not in the least his ability to traverse boundaries between different each challenging areas of physics and physical chemistry, he has left an immense and lasting impact on the physical, mechanical and chemical sciences.

As guest editors of this special 100th V.G. Levich issue of Russian Journal of Electrochemistry we are grateful to the contributors to the special issue for their excellent papers and tribute to Veniamin Grigorievich Levich. Continuous support of the preparation of this special issue by the Editors of the journal has been of importance for successful completion of the special issue.

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