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REVIEW PAPER

Contributions of A.N. Frumkin and the Frumkin School to power sources research

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Abstract
The historical interplay between the theoretical researches of the Frumkin School and the development of modern power sources technology is described. We show that the combination of solid fundamental knowledge with individual talent was the crucial factor in the development of power sources research and industry in the former USSR. Since historical trends tend to persist over time, it is clear that future developments of power sources will also require substantial inputs from basic electrochemistry, even if such requirements are not obvious to many present-day research managers.

Introduction

Although they had very different personalities, the names of Frumkin [1] and Bagotsky [2] continue to be very closely linked. Both played pivotal roles in the development of electrochemistry in the former USSR.

In the scientific literature, Bagotsky's name is most strongly associated with the development of power sources: he authored a number of well-known books and reviews in the area [3–9], and for many years this topic dominated his scientific output. By contrast, Frumkin's name tends to be associated with the double layer and electrode kinetics, and internationally he is regarded as one of the founders of "mercury electrochemistry." But despite their seemingly disparate specializations, both authors belonged to the same electrochemical community. They simply approached energy conversion problems from different angles (Fig. 1).

A graduate of the Frumkin School of interfacial electrochemistry, Bagotsky emerged as a leading figure of power sources research during the period of rapid development of electrochemical power sources in the post-war period. Amazingly, only five years passed between the 1952 textbook of Frumkin, Bagotsky et al. [10] (which omitted discussion of power sources altogether), and the 1957 launch of the "Sputnik" satellite containing silver–zinc batteries made under Bagotsky's supervision. This remarkable progress was made possible by the combination of Bagotsky's organizational skills together with his solid basic knowledge provided by the Frumkin School. This school, founded in the 1930s, generated many well-trained scientists for various areas of electrochemistry, including power sources.

The subject of "power sources" as an independent sub-field of electrochemistry has evolved gradually over two centuries of electrochemical development. But a key moment was the invention of the voltaic pile (galvanic cell), which provided a convenient and readily controllable source of dynamic electricity (1799). Initially, this device was indispensable for electrochemical experiments. However, in the decades that followed, several other power sources were also developed to supply the emerging industries of electrolysis, electrowinning, and electroplating. These included the *Daniell cell* (1836), the *Grove cell* (1839), and the *Leclanché cell* (1866). A clear account of these developments was published by Frumkin in his preface to the 1957 Russian translation of Jacobi's works [11]. However, as the number of industrial applications increased, and the variety of scientific phenomena expanded, some

Dedication We dedicate this article to the memory of Alexander Naumovich Frumkin (1895–1976) and Vladimir Sergeevich Bagotsky (1920–2012). We also celebrate Bagotsky's centenary in 2020.

The first author of this article has spent the majority of his scientific career in power sources research. In contrast, the second author has always avoided this topic. Nevertheless, they share an interest in the history of electrochemistry as a professional hobby.

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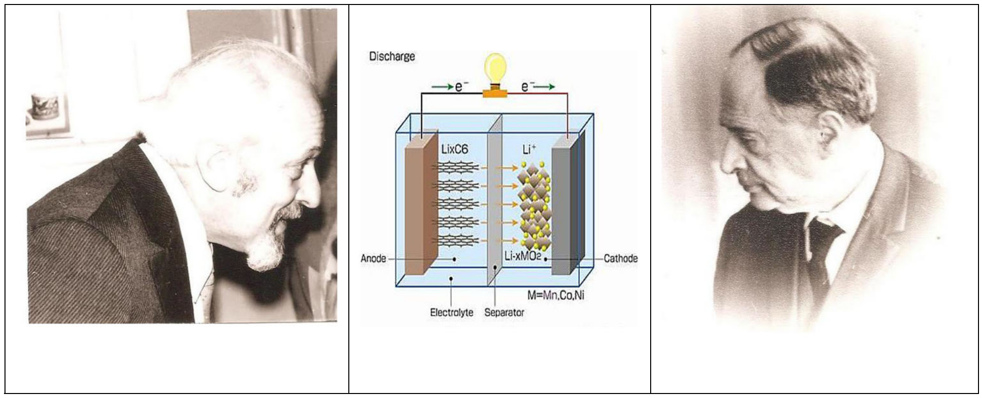

Fig. 1 A.N. Frumkin (at the right) and V.S. Bagotsky (at the left). We arranged these portraits around a sketch of the lithium-ion battery because the expressions accurately reflect their attitudes to power sources research

specialization among electrochemists became unavoidable. As a result, a certain narrowing of outlook became noticeable after the late nineteenth century.

Frumkin initially focused on interfacial phenomena, the importance of which was scarcely recognized in the 1910s–1920s. His studies at clean and well-defined mercury surfaces [12–14] resulted in the confirmation of various thermodynamic relationships between electrode charge and surface tension. Frumkin's methods were so admired that they soon became the paradigm of international efforts in colloid science and thermodynamics, and slightly later in electrochemistry as well.

Frumkin operated at the junction of several major disciplines within physical chemistry. This allowed him to make fundamental contributions across a wide range of specializations. He was a true *physical-chemist*. However, throughout his whole life, he considered himself a *classical electrochemist* and an apologist for electrochemistry. In particular, he facilitated the organization of a number of specialized electrochemical research organizations. He became the Head of the Laboratory of Technical Electrochemistry at Moscow State University (MSU) in 1930, then founded the Department of Electrochemistry at MSU and headed this Department from 1933 until 1976. He also initiated the Division of Electrochemistry in the Institute of Colloid Chemistry and Electrochemistry (named the Institute of Physical Chemistry starting from 1945), where he was also the Director (from 1939 to 1949). In 1957, Frumkin's Division of Electrochemistry became a separate Institute of Electrochemistry of the Academy of Sciences USSR, and Frumkin was appointed its first director. This Institute (now merged with another one) is widely known as *Frumkin Institute*. In 1965, to his great credit, he also launched the specialist journal *Elektrokhimiya*, English-language versions of which are known as *Soviet Electrochemistry* and, latterly, the *Russian Journal of Electrochemistry*.

Although he recognized the importance of applied electrochemistry, Frumkin was nevertheless more inclined towards basic research and the development of the philosophy of electrochemistry. In what follows, we describe a number of Frumkin's fundamental ideas which had a significant impact on the development of power sources. In particular, we single out (i) the solution to the "Volta problem," which was solved by introducing the concept of a potential of zero charge; (ii) the development of the "slow discharge" theory which addressed double layer effects on the rate of electron transfer; and (iii) the proposal of the Frumkin isotherm, which is still used today in intercalation systems. Besides these signature achievements, Frumkin made important contributions to the study of (iv) particular electrode materials and processes related to power sources and (v) porous electrode theory.

Frumkin's ideas and activity in the field of electrochemical power sources

Problem (i) was addressed by Frumkin at the very beginning of his career. The **Volta problem** had arisen soon after the first voltaic cell appeared. It amounted to two questions concerning voltaic cells: (1) how is the energy stored? and (2) where is the *emf* located? Both questions were the subject of heated discussions throughout the nineteenth century (ref. [11] contains a number of examples). The first question was answered by Michael Faraday in 1834 [15], and confirmed by Nernst in 1889 [16]; the storage of electrical energy occurred at the electrode/solution interfaces. However, the second question remained open for a much longer time.

On closed circuit, a typical voltaic cell has four phase boundaries which may contribute to the observed *emf*. These are the two internal electrode/solution interfaces, an external metal/metal interface between the electrode connections, and a liquid junction between the solutions. According to Volta and his supporters, the *emf* of this arrangement was primarily generated at the external metal/metal interface. Such an interpretation (known as the "contact theory") was justified by an experiment known as the "main Volta experiment": when two different metals were brought into proximity in a vacuum, they developed a potential difference known as the "contact potential difference."

Proponents of the alternative "chemical theory" believed that the *emf* was generated at the electrode/solution interfaces. However, when Millikan [17] repeated Volta's experiments in 1921, and confirmed the existence of contact *emf* under conditions of high vacuum, everyone was left puzzled. But Frumkin (1928) found a way out. Stated in modern terms, Frumkin realized that electrons had a finite chemical potential inside the material phases, but not inside the vacuum. He then introduced the concept of a *potential of zero charge* to quantify the difference [18, 19]. To validate this approach, Frumkin extended his studies from mercury to gallium and amalgams, jointly with *Aleksandra Vladimirovna Gorodetzkaya* [18, 19]. The use of a non-standard *U*-shaped electrometer (Fig. 2) allowed the authors to obtain electrocapillary curves for liquid gallium at ~30 °C and to show that the difference of zero charge potentials of two metals was approximately equal to the difference in their electron work functions [18]. *Amalia Davydovna Obrucheva*, Frumkin's collaborator and wife, also contributed to this research [20]. The key concepts were summarized in a review [21] completed by Frumkin's close associate *Mikhail Isaakovich Temkin* [22, 23]. To introduce these new ideas to students, Frumkin also included his solution of the Volta problem in his 1933 translation of Eucken's textbook [24] (he also completely re-wrote some sections in Russian).

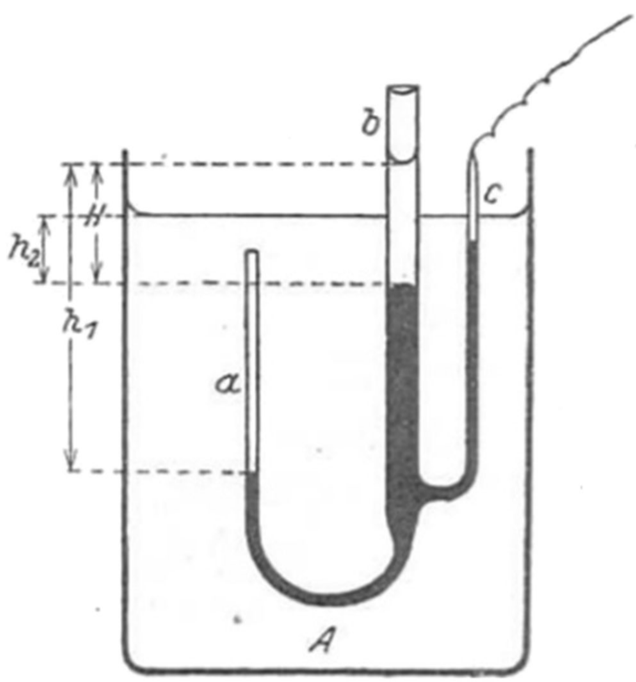

Fig. 2 U-shaped electrometer used by Frumkin and Gorodetskaya [18] to measure surface tension at liquid Ga/solution interface. Ga (black) occupies the asymmetric U-shaped vessel with different radii of the left and right tubes. The solution under study (A) contacts Ga in the narrower tube (**a**), and the reference solution (0.25 M KOH + 1 M KCl) is located above Ga in the wider tube (**b**). The additional tube (**c**) with a Pt wire serves for electric contact (potentials are the same at both interfaces). Surface tension can be calculated from H, h_1, h_2, radii of the tubes, and the tension at the reference interface

Problem (ii) is related to the **potential dependent rate of electron transfer**. This problem is much younger than problem (i). Indeed, the history of quantitative electrochemical kinetics is commonly assumed to start from the publication of the Tafel equation in 1905. Originally proposed for hydrogen evolution kinetics, this empirical relationship between current and overpotential was soon recognized as more-or-less universal for electrode processes. Indeed, it is difficult to overestimate the role of this equation in rationalizing the behavior of all types of power sources.

After a century of theoretical effort, the origin of the Tafel equation and its limitations are now clarified to some extent. The first milestone was reached in 1930, when Erdey-Grúz and Volmer applied a version of transition state theory to hydrogen evolution kinetics. By introducing a non-thermodynamic parameter α called the "transfer coefficient" [25], they obtained a compact equation for the current density, of the form:

$$i = k[\mathrm{H}^+]\exp(\alpha F\eta/RT) \tag{1}$$

Here i is the current density, k is a phenomenological rate constant, $[\mathrm{H}^+]$ is the reactant concentration, and η is the overpotential.

In the case of hydrogen evolution, the elementary act of electron transfer was considered to be the rate-determining step [25]. This contradicted Tafel's hypothesis that the slow recombination of two hydrogen atoms into one H_2 molecule was the rate-determining step. (Tafel thought that the electron transfer step was infinitely fast.) The possibility that electron transfer might actually occur at a low rate became known as the "slow discharge" concept.

Volmer's theory was able to explain the overpotential and temperature dependences of the current density but was still unable to explain the effects of ionic strength and the nature of the supporting electrolyte. In particular, salt additions to acidic solutions affected the rate of hydrogen evolution pronouncedly. In the theory-building work [25], it was tacitly assumed that the interfacial concentration of hydrogen ions was the same as its bulk concentration. Frumkin's 1932–1933 idea was to account for the fact that the discharge of hydrogen ions takes place inside a double layer. To capture this idea quantitatively, Frumkin turned to the Gouy-Chapman-Stern formulation of the double layer and expressed the surface concentration of hydrogen ions $[\mathrm{H}^+]_s$ as follows [26]:

$$[\mathrm{H}^+]_s = [\mathrm{H}^+]\exp(-\psi_1 F/RT) \tag{2}$$

where ψ_1 is the potential drop across the diffuse part of the double layer. In an earlier version of his theory, Frumkin had used the ζ-potential originating from colloid chemistry. Later the ψ_1-potential was used, as being the better parameter for quantifying the location of the reactant at its closest approach. In addition, Frumkin accounted for the fact that the driving force of the electrode process was not the entire potential E, but only that portion which corresponded to the dense part of the double layer. As a result, Eq. (1) was converted to the following equation:

$$i = k[\mathrm{H}^+]\exp(-\psi_1 F/RT)\exp(-\alpha F(E-\psi_1)/RT) \tag{3}$$

where E is the electrode potential. Today, the approach based on the analysis of polarization curves within the context of Eq. (3) is widely referred to as the "Frumkin correction." The novelty of Eq. (3) consists in the application of classical electrostatics to electrochemical kinetics. Even though transition state theory has today been eclipsed by more advanced theories, electrostatic effects on electron transfer remain an active area of electrochemical research. These effects sometimes allow one to change the electron transfer rate by orders of magnitude simply by changing the supporting electrolyte.

In passing, it should be noted that some corroborative evidence of the slow discharge theory was obtained by the Frumkin School as early as 1940, when accurate measurements were made of the frequency dependence of the capacity of platinum electrodes in the presence of adsorbed hydrogen [27, 28]. In this respect, considerable credit also belongs to Frumkin's collaborator, *Boris Vladimirovich (Wulfovich) Ershler*.

One of the most famous of Frumkin's theoretical constructs was (iii) an adsorption isotherm accounting for lateral interactions between adsorbates [29]. This isotherm was his first (and very successful) foray beyond thermodynamics, and it remains important in interfacial electrochemistry to this day. The role of the Frumkin isotherm in modern power sources is very important: it allows one to separate intercalate-lattice and intercalate-intercalate interactions [30].

In its earliest manifestation [29], Frumkin's isotherm took the form:

$$Bc = [x/(1-x)]\ \exp(-2ax) \tag{4}$$

where B is a constant dependent on the adsorbate-support interaction energy, c is the adsorbate concentration in the bulk of solution, x is the fractional surface coverage (θ in later works), and a is the lateral interaction constant. According to this equation, positive values of a correspond to attraction. In later forms of this equation, one also finds the parameter $g = 2a$ or even $g = -2a$ depending on the definitions of terms.

The solution of the Volta problem, the development of electrode kinetics, and the introduction of the Frumkin isotherm were truly landmark achievements. They placed Frumkin in the first rank of world electrochemists and established his scientific credentials in his own country. In retrospect, it is clear that Frumkin's personality was also very important: it acted like a magnet that pulled talented scientists into his orbit. By the mid-nineteen twenties, the stage was set for the rapid growth of a new scientific school.

Power sources advances in the first generation of the Frumkin School

In 1922, soon after Frumkin completed his pioneering thermodynamic studies [14] in Odessa, he moved to the Karpov Institute in Moscow. The "Frumkin School" emerged from that Institute, which was one of the first research institutions in the Soviet Union. The availability of talented collaborators allowed him to address a wide range of electrochemical topics, and in particular to advance on a broad front in the study of materials which later became very important for power sources. For example, in the early stages of Frumkin's work in Moscow, adsorption on activated carbon was addressed (with A.A. Donde) [31]. It was found that activated carbon in contact with air adsorbed hydrogen ions from aqueous solutions, whereas carbon saturated with hydrogen gas adsorbed hydroxide ions. In an extensive series of studies (e.g., [32–35]), Frumkin's team thoroughly investigated these phenomena and by these means opened direction (iv). Frumkin's young collaborators came from diverse backgrounds, and all arrived with independent research experience. *Natalya Alekseevna Bach* (the daughter of Alexey Nikolaevich Bakh, the founder and first director of the Karpov Institute) moved from Geneva. *Rebekka Khaimovna Burshtein* was a graduate of Leningrad University, while *Serafima Davydovna Levina* and *Boris Pavlovich Bruns* both graduated from the Kiev Polytechnic Institute. Frumkin co-opted all of them for his studies of the electrode/solution interface.

Before World War II, almost all scientific research in the Soviet Union had a defined military goal. Given the importance of activated carbon as an adsorbent, Natalya Bach worked hard to improve the synthesis of various carbons and to study the mechanism of their activation. She and Boris Bruns organized an industrial-oriented laboratory for their studies of activated carbon. One of the problems they solved was the limited performance of Leclanché cells, the most common power source at the time. The discharge of these cells depended strongly on the nature of the carbon components in the positive electrode. Natalya Bach also demonstrated the possibility of using domestic carbon instead of relying on imported carbon. This work had a pronounced industrial character[1]. Surprisingly, given the fundamental focus of Frumkin himself, his employees somehow managed to avoid becoming bogged down in the vexing problems associated with the

[1] We should mention that one of the motivations for the studies of surface phenomena was initially mineral flotation, a very important technological process in the field of hydrometallurgy [36].

kinetics of Leclanché cells, which was happening overseas during the same period (see, e.g., [37, 38]).

In 1930, after graduating from MSU, *Boris Nikolayevich Kabanov* joined the Karpov Institute. After a short period spent in the galvanics laboratory of D.V. Stepanov, he moved to Frumkin's lab, where he became one of the key figures. At MSU, Frumkin also found another important associate, *Zinovy Aleksandrovich Iofa* (Fig. 3), who had acquired electrochemical expertise from prior work with Evgeny Ivanovich Spitalsky. Kabanov and Iofa were very actively involved in the application of electrode kinetics to industrial problems. For example, Kabanov studied the kinetics of hydrogen evolution at ultrahigh current densities with respect to bubble formation [39]. Likewise, Iofa dealt with hydrogen overvoltage in various electrolyte solutions, particularly at high concentrations [40]. This was an extension of his work on the slow discharge theory (his DSc thesis on this topic was defended in January 1941). These studies assumed active interplay of electrode kinetics and interfacial thermodynamics.

In parallel, both Iofa and Kabanov were dealing with applied research, especially the lead-acid battery [41–43]. In line with global trends of that period, the key aspects of this research were more or less technical (such as the corrosion of positive plate grids, and the mechanism of expander action). Although these battery topics were not so interesting for Frumkin, he must have been interested in the hydrogen evolution reaction on lead, since the dependence on metal composition formed a special branch of his slow discharge theory.

Much research on the lead-acid battery industry remained unpublished. For example, it is only from the archives of Moscow University that we know that Iofa developed a molten salt electrolysis technique for the fabrication of Pb-Ca alloys, to replace pure lead. In co-operation with another of Frumkin's collaborators in Moscow University, *Mikhail Abramovich Gerovich*, Iofa also carried out lead-acid battery research in Ashkhabad (Turkmenistan) during WWII, as evidenced by the records of awards. Later, Iofa focused on corrosion research, although he also participated in a number of studies of air-polarized electrodes [44–46].

Fig. 3 Zinovy Alexandrovich Iofa in the laboratory of electrochemistry, Moscow University (old building), 1930s. From the private collection of M.Z. Iofa

Kabanov continued with lead-acid battery research throughout his life, pioneering the study of lead sulfate crystallization on lead [43]. He also developed various standards for improving the manufacture of lead-acid batteries, and published a number of fundamental works on the electrodes of these batteries [47, 48]. Today, Kabanov is also known for his battery-related studies of electrode processes on iron [49, 50], zinc [51], and magnesium [52], as well as silver oxide electrodes.

One more graduate of Moscow State University, *Nina Alexandrovna Aladjalova*, should be mentioned for her contributions to the study of electrochemical intercalation [53]. The particular system which she studied jointly with Frumkin was palladium-hydrogen, which was far from being a practical power source, but which nevertheless was a very useful model system for intercalation reactions coupled with ion discharge. The Frumkin-Aladjalova study contained two important innovations. First, their method of studying the content of hydrogen in the PdH_x hydride ("equilibrium charging curves") was a direct predecessor of the galvanostatic titration method widely used today. Second, the overall approach they applied to PdH_x (later developed further by Frumkin and co-authors [54, 55]) forms the basis of modern studies of metal hydride batteries. Indeed, the electrochemistry of hydride-forming intermetallics was systematically addressed by the second generation of the Frumkin School in the 1980s–1990s [56, 57].

Another intercalation system, the nickel oxide electrode, was studied by B.V. Ershler and his collaborators [58, 59]. This may be surprising to younger electrochemists, who generally associate Ershler's name with his famous equivalent circuit for impedance studies. However, Ershler, with his engineering background from Moscow Higher Technical School, actually performed many applied studies. We might also mention Kabanov's brief contribution to intercalation in the 1960s, namely his observation of alkali metal insertion into solid metallic electrodes from aqueous solutions, with formation of solid solutions and intermetallics [60].

In the aftermath of WWII, N.A. Bach, B.P. Bruns, and B.V. Ershler were compelled to change their scientific areas, but all were successful in their new fields. By contrast, S.D. Levina, R.Kh. Burshtein, B.N. Kabanov, and Z.A. Iofa managed to spend all of their scientific lives in the vicinity of Frumkin (Iofa at MSU, the others at the Institute of Electrochemistry). Probably the most systematic (although least known) series of studies related to power sources was that due to Burshtein. She developed activated carbons for electrodes with air depolarization. Also, when the production of Leclanché cells encountered a shortage of raw materials, Burshtein proposed the cheaper (albeit less efficient) iron-carbon cell. The negative electrode was made of scrap iron, and the positive electrode was made of carbon.

The engineer *Pavel Markovich Spiridonov* also worked along similar lines (creating air depolarization cells), becoming a member of the Frumkin lab at the Karpov Institute after graduating from the Mendeleev Institute of Chemical Technology. In 1941, Spiridonov was awarded the highest Soviet award ("Stalin Prize") for inventing a new type of air depolarization cell. However, even more notable was Spiridonov's oxygen-hydrogen aqueous fuel cell (formerly called the "gas cell") with gas-diffusion electrodes. He achieved current densities of 30 mA/cm^2 [61], which was a remarkable achievement in that period of exclusively high-temperature devices (F. Bacon's epochal work on aqueous fuel cells was not completed until the 1950s).

Many design concepts of modern fuel cells originated in Spiridonov's research: the filter-press design of multi-cell stacks, gas diffusion electrodes in contact with an electrolyte solution on one side and a gas on the other, and a catalyst layer located in the active zone of the electrodes. Later, Spiridonov also worked on the iron-carbon cell proposed by R. Burshtein [62], and on other cells with air electrodes. From her side, Burshtein extended activation carbon studies to platinized carbon [63], forming a solid basis for later studies of fuel cells.

As for Frumkin's personal attitude towards this work, he seems to have been a keen observer, always on the look-out for new fundamental problems. A good example is his interest in diffusion in porous systems, which opened direction (v). The electrodes of power sources are often porous and filled with electrolyte solutions. The resulting behavior is then spatially non-homogeneous, and requires special mathematical treatment because the systems contain distributed parameters. In particular, the rate of current-producing processes in the depth of the pores is less than the rate of current-producing processes in the mouths of the pores. Frumkin was one of the first electrochemists to replace the full 3D problem with its one-dimensional analog: his calculation of the current distribution along a single tubular pore [64] generated results having both theoretical and practical value. He considered the limiting cases of low and high polarization (the linear and exponential regions of the current density, respectively).

The second generation of the Frumkin School (Bagotsky period)

When identifying members of the first and the second generations of the Frumkin School, we take into account their age difference and (more importantly) the difference in preceding education and experience. The first generation of the Frumkin School consisted of individuals trained in different scientific traditions. By contrast, the second generation consisted largely of electrochemists educated within the Frumkin School itself (starting from their MSc or PhD period). This specialized electrochemical education was available at MSU even before WWII, and many young people joined in 1940s (Fig. 4). PhD positions were also created in the Frumkin's labs at the Karpov Institute and later in the Institutes of the Russian Academy of Sciences. These were very helpful in attracting graduates of technological universities.

The closest pupil of Frumkin, and the outstanding representative of the second generation, was *Vladimir Sergeyevich Bagotsky*, who graduated from Frumkin's Department at MSU in 1944, and spent about two years there after his PhD (completed in 1947). At that time, some staff members in the Department of Electrochemistry were investigating the oxygen reduction reaction on carbon [44, 45]. Bagotsky carried out a fundamental study of this reaction on mercury [65, 66], and also published results on other electrode materials. His coauthor in these studies was PhD student *Irina Evgenievna Yablokova*, who became his close collaborator, friend, and wife for more than 60 years.

Political circumstances prevented Bagotsky from working directly with Frumkin during the terrible period of the "anticosmopolitan campaign" (1948–1953). In 1949, he moved to the Research Institute "Cells-Electrocarbon"[2], later known as the All-Union Scientific-Research Institute for Power Sources (Russian abbreviation VNIIT). It was hardly a free choice, but it was a lucky one for the development of electrochemical power sources in the USSR. By 1951, Bagotsky had become the head of the Laboratory for Novel Electrochemical Systems in VNIIT. The area into which Bagotsky was plunged was the development of power sources for the military: mercury-zinc cells, water activated reserve cells with magnesium anodes, and thermal reserve cells for rocket technology. The most important work carried out at the Institute of Power Sources was the development of silver-zinc batteries, initially intended for jet aviation (MIG aircraft) and for submarines, but later for space technology. The launch of the first artificial satellite in 1957 and then the first manned spacecraft in 1961 were the most impressive consequences of the development of silver-zinc batteries under Bagotsky's supervision.

Historically, one must distinguish between the batteries produced in pre-revolutionary Russia (typically Leclanché cells and lead-acid batteries) and those produced for military applications after WWII (such as mercury-zinc primary cells and silver-zinc secondary cells). For the older group of devices, electrochemical research was mainly aimed at particular improvements in performance. As for the newer group, the technology had to be developed from scratch. Although none of these batteries was actually invented in the USSR (the prototypes were based on overseas patents), many scientific studies were required to obtain stable operation and to understand the essence of the electrode processes. Very little of this work

[2] Institute name in Russian was "Elementno-elektrougol'nyi institut."

Fig. 4 Department of Electrochemistry, Moscow University. Sitting (from left to right): Z.A. Iofa, A.N. Frumkin, M.A. Gerovich. Standing behind them (from left to right): A.D. Obrucheva, N.B. Moiseeva, A.I. Fedorova, O.L. Kaptsan (later Kabanova), S.Ya. Mirlina. In the back (from left to right): N.V. Nikolaeva (later Fedorovich), R.I. Kaganovich, V.S. Bagotsky, V.A. Kuznetsov. The end of the 1940s. From the collection of the Department of Electrochemistry, MSU

was published. However, a few articles did manage to escape censorship [67–74], and these happily preserve the names of many of Bagotsky's colleagues in that period (though not all). In this regard, *Iosif Arnol'dovich Zaidenman* deserves special mention, as someone who played a key role in the development of thermal reserve batteries. Bagotsky's lab in VNIIT became a lively center not only for battery technologies but also for the training of power source researchers. In this lab, former MSU graduates mingled with graduates from more technological universities, who introduced a strong engineering component into joint activities.

In 1959–1960, Frumkin, observing the development of fuel cells in the USA, the UK, and other Western countries, initiated a fuel cell program in the Soviet Union. Bagotsky was appointed as one of the leaders of this large project (more than a dozen research and industrial organizations participated) and also became a part-time researcher in the Frumkin Institute starting from 1960. The work program encompassed several different types of fuel cell, and almost all aspects of the theory. As it turned out, the take-up of fuel cells in the USSR was rather limited, but the fundamental research brought rich dividends related to the theory of porous electrodes (and, in general, of systems with distributed parameters) and to the kinetics of electrocatalytic processes. Building on the pioneering work of Frumkin [64], complex theories of liquid porous electrodes [9] and gas diffusion electrodes [75] were developed.

Fruitful ideas about bi-porous gas diffusion electrodes [76] were also suggested by Alexander G. Pshenichnikov, a pupil of Frumkin and Burshtein. In contrast to Bacon's bi-porous systems (in which a fine-dispersion layer plays the role of a gas-tight layer, preventing gas leakage into the electrolyte chamber), Pshenichnikov developed a bi-porous active layer containing gas-filled "wide" pores and liquid-filled "narrow" pores. The intersection of these pores of different sizes created three-phase boundaries at which the current-producing reaction occurred. Experimentally, porous electrode problems were tackled in a style very similar to that of the Frumkin School. Indeed, one can readily observe the similarity between the new cell designs (see Fig. 5) and the older "mercury era" cells.

Theoreticians also played an important role in this story. The central figure in this regard was *Veniamin (Benjamin) Grigorievich Levich* [77], who in the early 1940s became an associate of Frumkin in the Institute of Colloid Chemistry and Electrochemistry. Levich was a former student of the world-famous theoretical physicist, Lev Landau, and Levich's 1952 textbook entitled *Physicochemical Hydrodynamics* has become an international classic. The *Levich equation*, which describes the limiting current at a rotating disk electrode, is named after him. His research interests also extended to the quantum mechanics of electron transfer. Levich headed the Theoretical Division of the Frumkin Institute (an unusual arrangement in chemistry-oriented institutes). Bagotsky's division, launched in 1966, was the power sources capital of the Frumkin Institute, attracting people and ideas from various other divisions. Joint publications between theoreticians and experimentalists demonstrate their very close cooperation.

Fuel cell research required a deep understanding of electrocatalytic processes. In the 1960s, the oxygen reduction reaction and the oxidation of small organic molecules became topics of intense fundamental interest. Multiple studies of the oxygen reduction reaction were reported by Bagotsky, Burshtein, and their various pupils (see, e.g., [78–83]), while the oxidation reactions of methanol and some C2 species were of interest to Frumkin, who involved people from the MSU branch of his school to these studies [84–86]. It is easy to see the link between these works and Frumkin's early

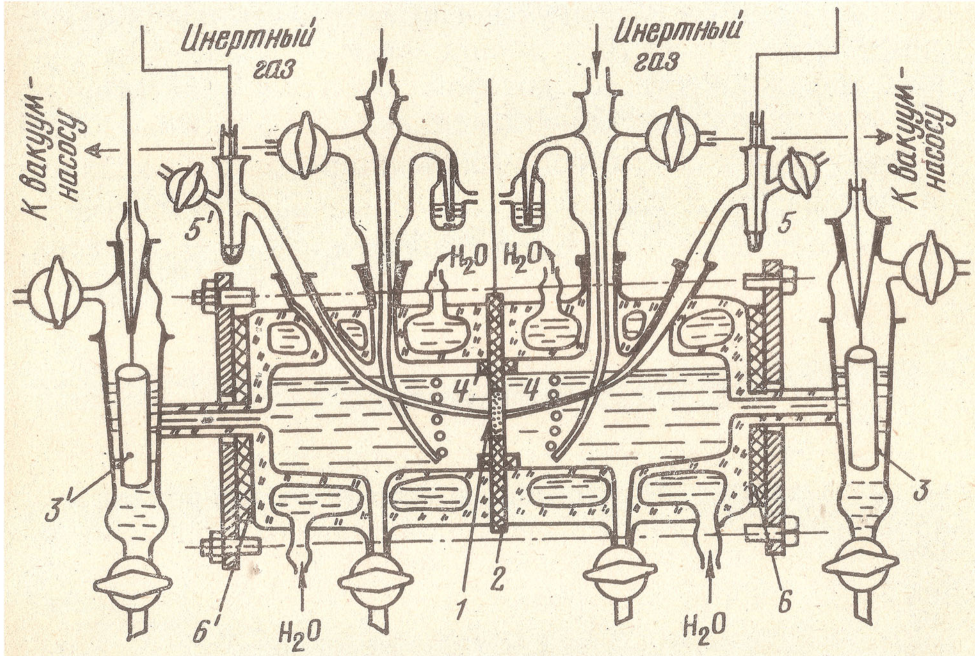

Fig. 5 The cell for measuring effective resistance. (1) porous electrode clamped between two cell sections; (2) PTFE sleeve; (3, 3′) counter electrodes in separated compartments; (4, 4′) Luggin capillaries; (5, 5′) reference electrodes; (6, 6′) rubber seals. Deaeration of solutions with inert gas, vacuum pumping, and water-jacket are envisaged. Reproduced from [9]

research on the adsorption on platinum group metals [87–89], especially in respect to experimental techniques. Active studies of electrocatalysis were also associated with Bagotsky's close collaborator *Yury Borisovich Vassiliev* (Fig. 6), whose lab in Bagotsky's division also produced many well-trained electrochemists (see, e.g., [90–96]).

Towards the end of Frumkin's life, power sources with lithium anodes became a hot topic. However, the high reactivity of lithium metal made cells of this type very difficult to assemble. Frumkin himself was never engaged in such experiments, but his intuitive feeling was that something revolutionary was about to happen in the lithium battery community. As Head of the Institute, he strongly supported research on lithium cells in Bagotsky's division. A special lithium research group was created, and also the efforts of researchers at other centers were welcomed. Frumkin also initiated a number of basic studies of non-aqueous electrochemistry at his department in MSU. After Frumkin's death, research into the development of lithium primary cells continued with the active participation of Bagotsky, and some lithium-ion batteries were studied too [97–99].

Finally, it would be remiss of the authors not to mention two graduates of Frumkin in MSU. These are Yurii M. Povarov who completed the first (1983) "lithium" book in Russian [100] and Valeriya S. Dubasova who developed lithium primary cells at her institute NIIEI[3]. In collaboration with Bagotsky's division, she also worked extensively on lithium-ion batteries [101]. Figure 7 shows the symbolic lithium gifted to Bagotsky on occasion of his 60th birthday.

Concluding remarks

The evolution of a scientific school is always a complex and dynamic phenomenon. It involves an intellectual tradition and a specific style of research, including very tiny experimental details. An important signature of a real school is its noticeable

Fig. 6 Yury B. Vassiliev, close collaborator of Bagotsky in the field of electrocatalysis

Fig. 7 Celebration of Bagotsky's 60th birthday (1980). Yu.M. Povarov, the head of the lithium systems group (on the left) presents the symbolic Li element (surely made from a less active metal) to Bagotsky (on the right)

diversity from other groups of researchers working in the same field. The success of the Frumkin School resulted, in part, from its openness to people with a wide variety of backgrounds and different vivid personalities. However, the spine and brain of this school were also continuously invigorated by Frumkin's fruitful ideas, which were always broader than any particular problem under consideration.

In the small space available in this document, it has not been possible to mention the names of all the school members who ever collaborated with Frumkin. Instead, we have deliberately restricted our attention to just one branch of a multi-branched research tree, which is often obscured by other branches associated with Frumkin's name. In our estimation, at least 30 MSU graduates worked in the field of power sources, a number comparable with those receiving an electrochemical engineering education. According to the folklore of the Frumkin Institute, the MSU graduates were always able to explain why certain devices failed, while the members with more technological education were able to make devices that worked, but could not explain how they did it. This may indeed be a Universal Law.

Of course, Russian power source developments were not confined to the Frumkin School. Two specialized departments of electrochemistry were established in addition to Frumkin's department in MSU: one in Leningrad (now St. Petersburg) in 1940 and one in Rostov-on-Don in 1976. There are also several dozen electrochemical engineering departments in Russian technical universities. Contemporary with the Institute of Electrochemistry in Moscow, the Institute of High-Temperature Electrochemistry was established east of the Ural Mountains (in Sverdlovsk, now Yekaterinburg). During the Frumkin period, this institute was headed by *Sergey Vasilievich Karpachev* (Fig. 8). Although Frumkin himself was never engaged in electrochemical experiments at high temperatures, he actively supported the work of his Ural colleagues. They studied zero charge potentials in molten electrolytes and developed high-temperature electrochemical devices. In this respect, we should mention *Mikhail Vassilievch Perfiliev* (solid electrolytes, the first soviet 1 kW SOFC battery), *Sergey Fedorovich Pal'guev* (basic studies of solid electrolytes for fuel cells), and *Gennady Konstantinovich Stepanov* (the first soviet battery of molten carbonate fuel cells). One more important person, very close to Frumkin and his school, was *Evgeny Aleksandrovich Ukshe*, known

Fig. 8 Directors of two Institutes of Electrochemistry: A.N. Frumkin (on the left) and S.V. Karpachev (sitting on the right). Beginning of the 1970s

Fig. 9 A.M. Skundin (left) and V.S. Bagotsky. Celebration of Bagotsky's birthday (Moscow, 2000). Bagotsky departed for the USA in the 1990s, but continued to collaborate with his old department at the Frumkin Institute until the last moments of his life

for his contributions to both molten salt and solid electrolyte electrochemistry. In 1964, he created a specialized laboratory in Chernogolovka (Moscow region). Ukshe's works were of the utmost importance for SOFC development in Russia. We also commemorate *Oktavian Stanislavovich Ksenzhek* from Ukraine (Dnepropetrovsk, now Dnipro), whose works on porous electrodes were actively supported by Frumkin as early as the 1950s.

A separate strong applied branch of power sources research in Russia was located in Leningrad (currently St. Petersburg), where the battery institute had been established in 1924. The history of this branch requires a separate communication. *Sergey Georgievich Kotousov* from Leningrad was the originator of a soviet silver-zinc battery designed independently of Bagotsky's team. The most prominent Leningrad person in the power sources area was *Alexey Ivanovich Rusin*, the highest level expert in lead batteries. Although this branch hardly engaged with the Frumkin school, the competition was healthy and facilitated battery development.

Both Frumkin and Bagotsky contributed enormously to general electrochemical enlightenment. In respect to power sources, a number of monographs were published, as well as important translations [102, 103]. Nowadays one can even discern a third generation of the Frumkin School, spread over a vast geographical area. The contacts between this diaspora and the second generation still working in Russia always were (and still remain) very close. We hope to maintain this network for the benefit of all, and particularly for active international power sources research in the future (Fig. 9). Vladimir Sergeevich Bagotsky, with his unusual Swiss-Russian-American biography [2], remains a lasting symbol of international scientific collaboration, and a remarkable bridge between Frumkin's basic researches and modern electrochemical applications.

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