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A Biographical Memoir by
MICHAEL A. COPLAN

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BY MICHAEL A. COPLAN

RAYMOND MATTHEW FUOSS was born on September 28, 1905, in Bellwood, Pennsylvania, son of Jacob Fuoss and Berdie Zimmermann Fuoss. He attended Altoona High School in Altoona, Pennsylvania, which at the time was a thriving manufacturing and railroad center.

Fuoss entered Harvard University in 1922 at the age of seventeen. At Harvard his main interest was organic chemistry, but, while working as a private laboratory assistant for Professor G. S. Forbes the summer before his senior year, he developed an interest in electrochemistry. His first published paper, with Forbes and S. W. Glass, was on the topic of oxidation potentials and equilibria in the system chlorine, iodine, hydrochloric acid, and water. It appeared in the *Journal of the American Chemical Society* in 1925. A second paper with Forbes on the reaction of bromine and the chloride ion in hydrochloric acid appeared in the *Journal of the American Chemical Society* in 1927.

While at Harvard he studied mathematics with Coolidge and Birkhoff. Those were the days when every undergraduate was expected to take science and mathematics for each of the four undergraduate years. Fuoss also studied German, and this may very well have been the beginning of his lifelong interest in languages. It is interesting to speculate how Fuoss, a small energetic boy from an industrial city in central Pennsylvania, fit in at Harvard? Fuoss graduated from Harvard summa cum laude and Phi Beta Kappa in 1925 at the age of twenty after completing the four-year curriculum in three years. He seemed to have had an interest in football, at least as a spectator. He retained a loyalty for Harvard, participating in the small Harvard Club in New Haven during his Yale years.

From 1925 to 1926 Fuoss had a Sheldon Fellowship to study at the University of Munich. It was in Munich that he found his calling. He had a good opinion of his abilities in organic chemistry, as what summa cum laude Harvard graduate would not, but this changed in the course of his work with Wieland on the structure of the bile acids. His initial idea was to do Ph.D. research in organic chemistry, but exposure to lectures in thermodynamics and physical chemistry, along with some difficulty in finding the position of the double bond in the bile acids, caused him to change to physical chemistry. He attended lectures by Fajans on thermodynamics. Lange was also at Munich at the time. The only publication to come from the year in Munich was with Lange on the concentration dependence of the heat of precipitation of silver chloride.

Upon returning to the United States Fuoss married Rose Elizabeth Harrington. For one semester he was an Austin teaching fellow at Harvard in Chemistry B, and then in the winter of 1927 became a consulting chemist with the firm of Skinner, Sherman, and Esselen in Boston. He earned extra money by tutoring undergraduates evenings and weekends in a variety of subjects from chemistry to naval science. During this period his son, Raymond Matthew, Jr., was born only to die after two days of life. The consulting and tutoring brought him enough money so that by September 1930 he was able to resume his graduate studies.

Fuoss entered Brown University for his graduate work in order to study with Professor C. A. Kraus. Fuoss had ideas about organizing the large amount of data on the conductance of electrolytes in a variety of solvents, and Kraus was an inventive, original thinker. Scientific interest in electrolytes had been stimulated by the 1923 theory of Debye, but there seemed to be no systematic way of understanding the different experimental results. Kraus was willing to experiment with many different solvents and solutes and measured the properties of solvents as varied as liquid ammonia and liquid hydrogen cyanide. By astute choices of solvent, conductance data over a wide range of dielectric constant and viscosity were acquired. Conductance was also measured as a function of temperature.

Lars Onsager was at Brown at the time. Fuoss attended his lectures and this began an association that was to span more than thirty-five years. The 1932 paper that they wrote together on irreversible processes in electrolytes took up eighty-nine pages in the *Journal of Physical Chemistry* and remained the definitive treatment of the topic until it was taken up again by the two of them in the 1950s. Fuoss recognized that conductance for the wide variety of solvents and solutes for which there were data could be understood in terms of the electrostatic interactions between the ions in solution with the solvent taken as a continuum with the microscopic dielectric constant and viscosity taken to be equal to the macroscopic values.

The topic of Fuoss's Ph.D. thesis was the properties of electrolytes in non-aqueous solvents. It was completed in two years in 1932 under the direction of Onsager. The pace was accelerating. Fuoss was appointed research instructor at Brown in 1932 and later assistant professor for research, positions created by Kraus especially for Fuoss. Fully aware of Fuoss's abilities, Kraus encouraged him to broaden his education. Fuoss was sent to Michigan for a summer to attend the lectures of Sommerfeld and Pauli on quantum mechanics. In 1933 he took a leave of absence from Brown and, with an International Research Fellowship, returned to Germany to work with P. Debye in Leipzig for a year and with M. Wein for a short time at Jena. He also spent a summer with Fowler at Cambridge gaining experience with statistical mechanics. Quantum mechanics was not to play any role in Fuoss's research, but for many years he taught first-semester quantum mechanics for physical chemists at Yale.

Research during the Brown years resulted in thirty publications—many with Kraus. Several were published a number of years after Fuoss had left Brown. The papers are almost equally divided between experimental and theoretical research. The experimental work was mostly high precision conductance measurements of a wide variety of organic and inorganic salts in pure and mixed solvents. Of the irreversible processes in solution that can be measured, conductance can be done with the highest precision and provides the best test of theory. The goal was to collect data over a wide range of the parameters that affect conductance. Strong, moderate, and weak electrolytes were used, with the anions and cations chosen to cover a range of sizes and shapes. Solvents low to high dielectric constant and viscosity were used . Miscible mixtures provided the means for continuously varying the physical properties of the solvent. The work was characterized by careful attention to purity of the salts and solvents. Fuoss's experience with organic synthesis and purification was essential. The measurements themselves were of the highest precision attainable at the time and once again showed a meticulousness and attention to detail that was the mark of all of Fuoss's research. Temperatures were controlled to within 0.01°C; concentrations had to be determined to better than 0.1%; the conductance cells, many of original design, had to be stable; and the measurements themselves had to be done in a way that eliminated all electrode effects and avoided any heating of the solution.

The theoretical work was an extension of the Debye-Hückel-Onsager treatment of the conductance of strong electrolytes in high dielectric constant solvents. The publications were mostly on the topic of weak electrolytes and the relations between association constants and the electrostatic forces between the solute particles. There were also mathematical papers on the solution of the conductance equation and the evaluation of the constants, the limiting conductance Λ_o and association constant K_A. Because Fuoss had available his own conductance measurements, the comparison between theory and experiment was straightforward, with each new system providing both answers and questions that could only be answered by further experiments and refinement of the theory. Some of his papers published during the period are reviews of the progress that was made both with the experiments and theory. Exceptionally clear, these papers are both a compilation of the data and summary of the theoretical understanding of the experiments.

The research program that Fuoss undertook required experiments, calculations, and theory, all at the highest levels. Only a scientist who combined the skills of an organic chemist, a physical chemist, a theorist in continuum mechanics and a mathematician could hope to succeed. For Fuoss it was the ideal choice.

In 1935 The American Chemical Society presented Fuoss its award for promising young chemists. Fuoss was thirty at the time. In the spring of the same year his second child, Patricia Rose, was born, and he was contacted by the General Electric Research Laboratory, then and for many years to come the premier industrial research laboratory in the country. He spent the summer in Schenectady as a consultant. This was the middle of the Great Depression with no money available for university research. The General Electric Research Laboratory provided a level of equipment and support that no academic institution could match, and Fuoss made the most of the opportunity, joining the permanent staff of the laboratory the next year.

The situation with polymers at the time Fuoss joined General Electric was similar to that in electrochemistry several years earlier. There was a large amount of data on the electrical properties of polymers, but it was difficult to assess their quality; until this could be done there was little hope of correlating the composition of the polymer with its physical and electrical properties. By eliminating surface effects and controlling the ionic content of the polymers Fuoss was able to obtain consistent reproducible measurements. It then became clear that the dielectric response of polar polymers depended on the nature of the polar substituents and the degree to which they were able to follow a time-varying external electric field. The frequency region over which the dielectric properties of a given polar polymer changed was found to depend on the concentration of the plasticizer as well as the size and shape of the plasticizer molecules. Between 1937 and 1945 Fuoss published twenty-six papers on the electrical properties of solids, almost all of which were based on work done before the beginning of World War II; during the war he worked on classified research topics.

Once the war was over in 1945, Fuoss accepted the first Sterling professorship in the Department of Chemistry of Yale University. His qualifications were perfectly matched to the department at the time. At Yale were Lars Onsager and John Kirkwood, two of the world's leading theorists in statistical mechanics, both of whom had published important articles with Fuoss. Also there were Herbert Harned and Benton Owen, supremely talented experimentalists in electrochemistry. Andrew Patterson and Phillip Lyons would be added to the faculty a few years later. With Fuoss joining the faculty, the department could well be considered the best in country in the areas of electrolytes, polymers, and statistical mechanics research. Fuoss at age forty still had unlimited energy and had made important contributions in each of the areas. For Fuoss, after the war years, the appointment was the chance he was seeking to return to academic research, teaching, and the training of graduate students. Once again he would work on problems that interested him rather than those suggested by others. He was, however, neither impractical nor opposed to applied research. Industrial consulting was an important activity for him in the years after graduating from Harvard; at Yale he continued his contacts with industry by consulting for DuPont, Monsanto, the California Research Corporation, and Arthur D. Little.

Upon arriving at Yale, Fuoss took up a new line of research that drew on his experience with electrolytes and polymers. This research was concerned with the properties of polyelectrolytes, high polymers with positive charge sites all along the length of the polymer chain. The mutual repulsion of the positive charges could change the conformation of the polymer chain and dramatically alter the viscosity and conductivity of polyelectrolyte solutions as a function of solute concentration. Fuoss synthesized a number of polyelectrolytes and measured their properties in a variety of pure solvents and solvents to which simple electrolytes had been added. He was able to explain the results with a molecular model that took into account the structure of the polymer and the inter- and intramolecular electrostatic interactions. The similarity between polyelectrolytes, proteins, and membranes was obvious, but the way to apply the results of the polyelectrolyte research to proteins was not clear at the time. This research, continuing well into the 1950s, was gradually replaced by theoretical and experimental work on 1:1 electrolytes in a variety of solvents. In 1951 he was elected to the National Academy of Sciences.

With Onsager, Fuoss systematically reexamined the assumptions and approximations in the Debye-Hückel-Onsager theory of electrolytic conductance. By retaining higher order terms, Fuoss found that the ad hoc assumption of ionic association for electrolytes in low dielectric solvents was no longer needed. A term analogous to the ionic association term now appeared in the revised conductance equation. The price to be paid for this theoretical rigor was the appearance of a second parameter, the ion radius a_o, and a much more complex equation. The parameter a_o appeared both in the new ion association term and in one of the hydrodynamic terms. In principle, the values of a_o derived from both terms should have been the same. Whether this was the case depended on suitable experimental tests based on higher precision conductance data than ever before and a reliable numerical method for extracting the physical parameters from the data. With typical energy and planning Fuoss and his students constructed a conductance laboratory where measurements with a relative precision of 0.01% were routine. Data analysis was done on a series of Yale computers at the computer center. The new theory was not a complete success. For a large number of solute-solvent combinations the ionic radius derived from the hydrodynamic term in the conductance equation was essentially equal to the hydrodynamic radius, but for low dielectric constant solvents there were significant differences. Much of the remainder of Fuoss's scientific career was devoted to exploring the origin of these differences and seeking better approximations in the derivation of the conductance equation. In 1974 he retired from Yale University but continued active research in electrolytes. From 1974 to 1980, the date of his last publication, he published twenty-two articles in refereed journals.

The legacy of this phase of research is over eighty research and review papers that cover all aspects of electrolytic conductivity from the development of new instruments and techniques to high quality conductance data and the theoretical development of ever more refined conductance equations. In addition to the research papers, Fuoss wrote *Electrolytic Conductance* in 1959 with F. Accascina of the University of Rome. This small book of 279 pages is written in the Fuoss style: direct, clear, complete, and precise, with no gaps between equations to be filled in by the reader. Introductory chapters on hydrodynamics, statistical mechanics, thermodynamics, and distribution functions are excellent summaries that neither oversimplify nor confuse with excessive extraneous detail.

A great deal has been made of Fuoss's extraordinary facility with languages. He could read, speak, and understand nineteen languages; he spoke of his interest in foreign languages as an avocation or hobby, but went about learning them with the same resolve, energy, and organization that served him so well in research. He traveled widely and collaborated with scientists in several countries, always mastering the language of his host country. He lectured in French at the University of Paris, in Italian at the universities of Rome and Palermo, in Turkish at the Technical University of Ankara, and in Hebrew at the Weizman Institute and Hebrew University. This was as much an effort to understand what was going on around him as it was an interest in linguistics. Language, of course, requires one to mentally organize a large number of objects, words in this case, and find patterns. This was what Fuoss was best at in his research, and it is not surprising that he was able to apply his skills to this area. He saw his intellect as a tool to be applied to whatever problem was at hand.

Fuoss excelled at teaching and he enjoyed it. He routinely taught graduate courses in mathematics for physical chemists, quantum mechanics (essentially taking the viewpoint of applied mathematics), polymer chemistry, and electrochemistry. The lectures, planned with precision, contained no digressions; everything was well organized and clear. One came away from them, if not inspired, at the very least confident that the material could be mastered by diligent study; there were no mysteries nor imaginative leaps. In his research group organization prevailed and each member, whether an experienced research associate or first year graduate student, had a well defined project. Monthly reports, organized according to a strict format, were due from everyone the first Monday of each month. They were promptly read and returned with corrections. Three times per week there were group literature seminars at lunch. Although it never seemed so from the graduate student perspective, Fuoss was keenly aware of the abilities and limitations of the members of his group. This was reflected in the problems he gave them and the help he provided. Often impatient and abrupt, he was also encouraging and kind. His philosophy was that a research career was not made in graduate school but afterward. He provided the training and taught the skills; whether a student would go on to be a successful researcher depended on what was done after obtaining the Ph.D. degree. With the Fuoss system a Ph.D. routinely took three to four years. It is noteworthy that Fuoss himself finished his Ph.D. degree in two years. There was no spare time with this schedule, but the outcome was assured. A large fraction of former Fuoss students hold academic and research positions.

What kind of man was Raymond Fuoss? He was energetic, forceful, and impatient. There was no time to be lost. All routine tasks were accomplished on the spot at top speed to have time for the more important research tasks. He was single minded with respect to his work. While he would listen to others it was mostly to find the errors in their reasoning. He aggressively defended his work and his publications and was fair but uncompromising in assessing the work of others. As a self-made man he was a staunch conservative and an opponent of taxes.

He was devoted to his second wife, Ann Stein Fuoss, whom he married in 1947 after having been divorced from his first wife for some years. The Fuosses understood and complimented each other. Mrs. Fuoss provided a vivaciousness and ease that were appreciated by the professor. When Mrs. Fuoss died in 1979 it left a deep void in his life.

Raymond Fuoss left a number of contributions to chemistry and physics that are the basis of important fields of current research. He presented his conductance equation in a form that was accessible to experimentalists. He treated the hydrodynamics and electrostatics of ions in solution in a rigorous mathematical way and at the same time made high precision measurements to test the theoretical results. He took the initiative in providing experimenters the source code of the computer program used for the analysis of conductance. There were other theories at the time, but none in such a useful form. Fuoss recognized the biological implications of polyelectrolyte studies, and his formulation of the properties of polyelectrolytes remain a useful starting point for theoretical and experimental work on proteins. It is indeed rare to find a scientist with exceptional abilities in such a wide range of topics. Those who knew Raymond Fuoss appreciated his intelligence, hard work, and uncompromising devotion to scientific truth.

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