

An electrochemical life

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It was my aunt who started it. She had some iron filings on a piece of paper and a magnet underneath. When she moved the magnet, the iron filings moved. I was only 5 years old at the time, and I could not see any way that the magnet could have touched the iron filings.

Later on, in high school, I learned something about “lines of force” which were supposed to emanate from the north pole of a magnet, pass through surrounding space, and enter the south pole. The only problem with this idea was that I still could not see them; but I did learn the word “magnetism”, and I also started to collect notes based upon books that I read, and even encountered the phrase “magneto-chemistry”.

Many years later, I found myself at Imperial College in London, applying for a stipend to do research. I must have been about 20. The place was dark and deserted as I walked

hesitantly down the corridor, but there was a dim light glimmering under one particular door, so I knocked on it. The response was a military style “in”. The owner of the voice did not waste any time. He brusquely asked the purpose of my visit before saying “sit”. He then asked me several technical questions, of which I still remember three. The Third Law? Debye–Hückel Theory? Nitric oxide? The latter turned out to be an oblique query about *anesthetics*. He then asked me what kind of research I would like to do if I got a scholarship. When I told him that my greatest interest was in magneto-chemistry, his face fell. For my part, I had no idea I was talking to H.J.T. Ellingham, a world expert on the industrial applications of electrolysis, and, at night, a Lieutenant in the 2nd London Home Guard. He did not say anything. He just stood up and said “wait”.

About 5 min later he returned with a tall gentleman, who turned out to be H.J. Emeléus, the leading inorganic chemist in the country. His seminal book *Modern Aspects of Inorganic Chemistry* (1938), conceived and written with his colleague J.S. Anderson, had led to a national revival of interest in that area of chemistry. Unfortunately, Emeléus spoke very quietly, and I did not catch his name. So I did not realize to whom I was speaking. He came straight to the point.

“So you want to do magneto-chemistry?”

“Yes”, I said eagerly.

“Then I assume you know magnetism and electricity...?”

I nodded.

“Well, then, how about magneto-chemistry and electrochemistry...?”

I thought about it, but before I could muster a response, he continued, “If you’d like to do electrochemistry, we do have scholarships for that, but as for magneto-chemistry... the war is on and many of our people are in the army...” At that moment I knew I had become an electrochemist!

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A few weeks later, I was given a room (actually an office and a lab combined) and a pep-talk from the man who was going to be my supervisor, none other than Dr. Ellingham, the man to whom I had spoken on that first day. The pep-talk lasted about 20 min, and the only outcome of significance was that he handed me a box of abstract cards on which he had written the titles of interesting papers in electrochemistry that he had read over the previous few years.

We are talking about 1943 here. War conditions were chaotic and I was unclear how to proceed. All I had were the abstract cards and a few vague hints. Ellingham added, “I’m sure you will find something that has not been done yet... if you follow up an idea, and make something of it, you should be able to write a thesis and get a degree...” Ellingham was always brief, but 20 min hardly seemed adequate to map out the next 2 or 3 years of my life.

Anyway, I was left to my own devices, with the box of cards and a broad hint that non-aqueous solutions might be a reasonable place to start. I read, for about 3 weeks, digging out various references, reading the relevant papers. Suddenly, it hit me. *Overpotential had never been measured in non-aqueous solutions.*¹ Ah! That would be my project. If my way onwards was as open as it seemed to be, then I would set the direction myself, and here it was in work which had just been published, telling me where to go.

Well, what followed was 2 years of hard work, so I’ll spare you the minor details. But after 2 years, I had accumulated lots of data, though I certainly did not understand them all. One day, Dr. Emel us came to see me. As usual he spoke *sotto voce*. I was eating a sandwich at the time, when he suddenly whispered, “How long will it be, Bockris, before you can write your thesis?” I probably hummed and hawed a bit before trying to answer, because he did not wait for my reply, but continued, “...because if you can finish it in time to give the lectures which Ellingham was due to give before he leaves, we would probably give you a job on the faculty...”

Of course this really made me jump, because a faculty position at Imperial College was a prime job. It implicitly held tenure, and unless you punched your supervisor in the face or did something similarly outrageous, you were there for life. The only problem was that I had to finish my thesis before the lectures were due to begin, because, by

regulation, they could not have someone lecturing to students who was not fully qualified.

I wrote my thesis in 6 weeks, which included a nightmarish period of 3 days in which I did not sleep at all. But I managed to turn up at Ellingham’s door one Sunday morning, very cold, at the only time he had available, 7:00 a.m. I was to present him with everything I had done. He paced up and down while I spoke to him, and did not seem interested in the slightest. But he told me to leave the thesis with him and he would see what could be done. “Come back next Sunday morning at 7:00 a.m.”

Of course the week between the two meetings was one of extreme tension for me, as my future career was hanging in the balance. Then, at the appointed hour, pacing up and down before a blazing fire, he told me that he did not think the thesis was good enough. The obvious question was *why*? His answer, I was not prepared for. He said that there was far too much mathematics in it. I held precisely the reverse view, that unless your thesis had some mathematics in it, then it was not up to standard. He told me to “rewrite” it, before ushering me out into the cold street at about 7:20 a.m. I was now in suspense for another week.

To rewrite a thesis in a week is absurd, so I just rewrote the mathematical parts in plain English, indicating that they could be “supported” by mathematical equations. He was still doubtful the next week, but I explained that his departure for a job at the Royal Institute of Chemistry would leave a teaching gap in electrochemistry, and that I hoped to fill in. “If I don’t get the thesis sir, I cannot carry on your tradition...” At that point he softened somewhat, and said “I’ll let you go through to the oral, but the examiner may not let you through”.

I did not know who the examiner was at that time, but it turned out that he was a renowned surface chemist brought down from Edinburgh. Of course, I did a good deal of touching up of the thesis before the oral. When the examiner finally started the formal examination, he went more-or-less page by page, stopping and asking little questions, which I easily batted back. But I realized that I had made a big mistake slightly past the mid-point of the thesis, and that we were about to reach it. At that moment, the Great Hand from Above entered into my life, and the examiner suddenly said, “Well, that’s enough for today, Bockris, well done!” And that is how I became an electrochemist.

So, in September 1945, I began my career as a young academic at the Imperial College of Science and Technology, London University. I was only 22, which was very young indeed to have a position on the faculty. There was a good explanation for my youthful appointment, though. The war was still on, and many young people who would have been my competitors were away on active service. The following year, there was of course a flood of students

¹ I once set fire to a flask containing ether around 9:00 p.m. one evening. It did not seem to be the kind of thing that my supervisor would want to hear about, so I let it pass. By chance, seeing him in a queue at the hostel in which we both lived during the war, I boldly approached him hoping to get an appointment for a discussion of my work. When I got level with him in the queue and indicated that I would like to speak with him, he looked at me and said, “Is it about the theory of inflammability?” He did not give me an appointment.

who wanted to work in Universities. Among the throng were Brian Conway (who later emigrated to Ottawa, Canada) and Roger Parsons, who became famous in the academic world in England, ultimately becoming President of the Faraday Division of the Chemical Society, and a Fellow of the Royal Society.

The first time finance came into things was in 1947, when I was visited by two gentlemen from the British Iron and Steel Research Association. They came right to the point within 3 or 4 min of coming into my room. They wanted to know if slags were conductive. Unfortunately, all I knew about slags at that moment was that they were supposed to be very important in steel making! However, the models used by BISRA seemed to me to be oversimplistic (i.e. they were supposed to be molecular compounds and so should be non-conducting) and after some discussion they decided to fund some research with me.

Richardson was their research director and Jim Jeffs was his companion and—it seemed to me—Jeffs was the originator of most of the queries. I answered the most important question (are slags conducting?) within the first 6 months of the contract, by which time, of course, we had learned a lot about slags. The two researchers I supervised on slags were named Liebermann and Tomlinson. After we measured the conductance of the slags at the unheard of temperature of 1,850 °C, BISRA were quite keen to extend their funding to other measurements.

What surprised those with whom we spoke with about our work was the high temperature at which we worked, over 1,600 °C, which is normal in steel making but rather unusual in electrochemistry. I learned a lot about how to do this by visiting the steel makers of Britain at that time, and they willingly showed me their technology. The man who worked on the most difficult part of these measurements was called Dennis Lowe, and he had an unfortunate accident just as he was finishing the building of an extremely complex apparatus which rose about 12 ft above the ground. Putting the finishing touches to it, he stood upon a chair, lost his balance, fell over, and the entire apparatus (which had taken more than a month to build) crashed down on top of him. This did not put him off for too long, however, as he was soon back at it and performing accurate measurements of the viscosity of liquid silicates at temperatures above 1,800 °C.²

² Austen Angell is also familiar with this work which took place about 1950. He tells me that because he had to use data for very high-temperature liquids he became familiar with the details of the work we did years ago, and he felt in speaking recently, i.e. 2009, that even though more than 50 years had passed since we did this work, it still remained unsurpassed because of the high temperature at which we made the measurements.

When discussing those early years at Imperial College, the name of Roger Parsons inevitably turns up. Roger was an impressive student, and 60 years later I can still visualize his economy of movement. He would enter my room upon arrival every morning, and on cold days he would already be removing his overcoat with one arm as he entered. With the other arm, as it dropped down, he would be seeking to switch on his apparatus. Years later, Roger told me that he had chosen to carry out research under my direction because, whilst attending my lectures, he had noticed that I had delivered them without notes. He found this most unusual, and it had suggested to him that I must have known what I was talking about!

My own view of the young Roger Parsons was that he was an outstanding student. Indeed, his confidence and objectivity were so great that he would change his view—one theory, let us say, for another—without compunction, and without the least feeling of any great event having occurred.

When it came for Roger to get his Ph.D., and leave me, I felt that we had done a very important job for electrochemists in general, because we had actually found out how to get reproducible results. When we began, we would get certain data from certain experiments, and then go to other laboratories in England, and also some in Germany, and compare them. It was very disconcerting when we found that they were all different. But with plenty of help from Brian Conway, Roger found out how to purify solutions and electrode surfaces in such a way that the irreproducibility was overcome.

In the immediate post-war years, the University of Cambridge was the principal place in England at which first class electrochemical research was performed. The main person I knew there was John Agar. He stayed in Cambridge for his entire career, and was a leading light there for more than 30 years. T.P. (“Sam”) Hoar was another person I knew in Cambridge, and his *forte* was corrosion at a very fundamental level. In fact, in the history of this rather practical subject, Agar, Hoar, and U.R. Evans are generally credited with having established, for the first time, the basic mechanisms of this very important process. Perhaps they got some theoretical ideas from the publications of Karl Wagner, but it was Evans and Hoar who proved the mechanisms experimentally.

Hoar played a part in my life in the late 1940s because it was he who invited me to come with him to Brussels in Belgium where, he said, a very important meeting was to take place. I was uncertain as to what was going to happen there, until we arrived, when I was introduced to Marcel Pourbaix, a man throbbing with enthusiasm for his job which was of course the thermodynamics of corrosion. By the time that invitation

came I also knew perhaps 20 or 30 electrochemists in the USA, or at least I had met them.³

I have already mentioned Dennis Lowe as the man who measured the viscosity of silica at high temperature. It was with him that I developed the first theory of liquid silicates, because we also measured the viscosity of various mixtures of silica and metal oxides, and we decided to propose a fairly comprehensive theory of all kinds of silicates. This needs diagrams to express it properly, but I will just say that the principle anion was $\text{Si}_3\text{O}_8^{6-}$. The paper was accepted in the prestigious *Proceedings of the Royal Society*, which meant that it was very widely read.⁴

In 1953, I was appointed Professor of Physical Chemistry at the University of Pennsylvania. In 1956 Brian Conway, who had followed me to Pennsylvania, decided that the time was ripe to accept his own Professorship, and he took a position at the University of Ottawa, where he stayed for the rest of his life. However, we had a great collegueship which we managed to maintain for many years afterwards. With Conway I developed a good deal of my thinking on the hydrogen evolution reaction, which became the model I used for all other electrochemical reactions. Along with this, I was also interested in the oxygen reduction reaction (and its partner, the oxygen evolution reaction) because I was always interested in fuel cells.

Thinking about the evolution of oxygen puts me in mind of A.K.M. Shamsul Huq, who was a student who came over with me from London. Now, we had already established that impurities in solution were the main problem in getting reproducibility in electrochemical reactions, and so Huq and I were able to work at very low current densities or velocities of reaction. Eventually, we were able to make measurements as far down as 10^{-9} amps cm^{-2} . At the time we were doing this (1955), there was no air conditioning, except for apparatus. That is, the University would buy an air conditioner for an experimental piece of equipment, but not for a researcher!

³ In telling amusing stories which occurred on my long pathway through life, one has to be careful not to tell too many, because of course people get bored with them. However, I feel that this three- or four-liner will not bore you if I tell it, as it gets something of the atmosphere of the time and also tells something about how I was regarded by my students at this early time. The two young men who comprise this story lived together to save money, and they also slept in the same bed, particularly on cold days, for they could not afford any heating. On Saturday they would stay longer in bed, and on one occasion there was a ring at the doorbell and a telegram arrived. One of the boys got out of bed to fetch it, and read it to his startled companion upon his return. It said “The early bird gathers the worms. It’s already 9:30. Why aren’t you here? Bockris”

⁴ Proof of this statement came to me on the bullet train in Japan. I had never been on the bullet train before, and it was exciting to ride at 120 mph. However, the train does stop every now and again, and at one stop a man entered, came confidently up to me, and sat down. He explained that he had found out that I would be on the train, and wanted to congratulate me on my theory of “glasses”!

Researchers, they thought, should sweat it out, so we tended to work after dark when it was cooler.

Meanwhile, Conway and I continued to develop our theory of proton conduction. However, there came across our bow, so to speak, a man who later on became famous in his own right, namely Manfred Eigen, a German physical chemist. After we had obtained acceptance of our paper, in the *Journal of Chemical Physics* no less, he came out and said that one of the steps we had made was wrong in not being quantized when it should have been. Fortunately, this did not affect the overall model, which was that the rate determining step was related to the rotation of water molecules with which the proton was surrounded. Of course, the rate of rotation depended upon the electric field of the proton, and later on we showed why it was that, in ice, the proton had a much greater mobility than it had in water, a counter intuitive fact.

One of the more exciting students in the 1950s was a German called Wolfgang Mehl. The excitement he brought to our group consisted of two quite separate elements. First of all he brought from Berlin, where he had been working with a German electrochemist called Heinz Gerischer, the idea of “transients”, i.e. very fast measurements which could be over in a millisecond. There are various kinds of transients (do you keep the potential constant?) and with Mehl we developed several new ones. But, Wolfgang’s popularity and local fame among my students was more dependent upon his stories about the war.

C. Austen Angell worked with me in the 1950s too, and later on he had a splendid career, in what you might call metastable aqueous electrochemistry. His specialty was a subject called *supercooling*. He studied this phenomenon in great detail, and the results made him well known and the recipient of many awards.

In looking back over the years, one tends to pick out individuals of an unusual character, and one person who especially comes to mind in the late 1950s, was Erik A. Blomgren, a Swedish post-doc who was accompanied by his (unmarried) wife. He was in the habit of calling me up on the telephone when he had completed an experiment, and telling me about it by saying he had brought it “on the limit, John”, meaning that he brought it to an extreme never reached before!

Aleksandar (Asa) Despić was another man who worked with me in the 1950s. He also returned in the 1960s, on which occasion he was accompanied by his colleague Dragutin M. Drazic. These fellows were both strong communists. In those days in the USA, it was considered almost dangerous to admit that one had met a communist.⁵ Despić was particularly sought out by wealthy Americans,

⁵ When I was invited to Russia in 1956 to give a lecture in Moscow, I felt it prudent to approach the president of the University of Pennsylvania and ask his permission to accept the invitation, so great was the negative feeling towards communism at that time.

who wanted to meet an actual communist, and they were always surprised to find out that he was quite a normal and intellectual person. According to his own accounts, he actually had some success in bringing his hosts around to his way of thinking. He certainly left me with one memorable phrase from those days, “From each according to his ability, to each according to his needs”.

Despić and Drazic introduced me to the very important electrochemical process by which iron dissolves and is re-deposited on solid substrates. The reasons for my interest in this go back to my earlier discussions with Hoar and Wagner, both of whom I have already mentioned. What Despić and Drazic did was to explore the detailed mechanism of the iron dissolution reaction. This was important because of its engineering implications. Why do ships and planes not last more than 30 years, or so? It is all because of corrosion, of course, and I have heard it said that 2% or 3% of the GNP of the USA is still wasted on corrosion.

Before finishing this account of derring-do among experimentalists, I would like to briefly mention *dendrites*. In case you have not heard of them, they are spike-like things which jut out from growing bodies during crystallization. All this interested a man who came to work with me from England, whose name was J.L. Barton, and he resolved to find out how they worked. We published a paper together in the Proceedings of the Royal Society entitled “The electrolytic growth of dendrites from ionic solutions”, and that has become something of a classic. One of the more exciting mechanistic things we did was to find out why dendrites grow so fast. It turns out to be due to the spherical shape of the tip of the crystal. If the radius is less than the diffusion layer thickness then the rate of the growing crystal is dependent on R . If that is small enough, the dendrite grows so fast that it sticks out from the crystal body.

An electrochemical life

Part 2

1963 was a good year for me because it was the year in which I published my best paper. It was called “On the Structure of Charged Interfaces”, and was co-authored with M.A.V. Devanathan and K. Müller. It was published in the Proceedings of the Royal Society (London), and today is widely referred to as “The BDM Paper”. Devanathan was a post-doc with me, but he had also worked in London with Roger Parsons, and he was thoroughly knowledgeable concerning the structure of the double layer. Computers were just coming into university research in 1963, and in those days there was usually only one computer in a special

room, and one could only use it by appointment. I designated Müller to be the person to learn how to use it, and it was he who carried out all the calculations, of which this particular paper had many.

There was also a major innovation in this paper. It was the first time that the structure of water on the electrode was included in any model of the double layer. Obviously a layer of water dipoles would be oriented according to the charge on the electrode surface, and the extent of orientation would vary with the applied potential. The mathematical treatment of the water, and how we accommodated the potential of the electrode, was largely developed by myself, but I must admit that I got a bit of help from a treatise on paramagnetism in which the orientation of the magnetic moments had been worked out earlier. Devanathan was responsible for most of the ideas on specific absorption, which also allowed us to interpret the characteristic “hump” on capacitance-potential diagrams, which had long puzzled researchers. We managed to get the BDM paper into the *Proceedings of the Royal Society* and it has been the “apple of my eye” ever since.

Things were going fast and furious in my group in the mid-1960s and it grew past 25 people. Eventually, we hired a man to look after the finances, although the University also kept its eye on that, but the details were very much in our own hands. Every year we looked forwards to determining the number of coworkers we could hire.

Butler story 1

In order to understand this story, one has to have some idea of the era to which I refer. It was the early 1950s. At that time, J.A.V. Butler was working at the Chester Beatty Research Institute, Institute of Cancer Research, which had its building remarkably close to the Imperial College building in which I worked from 1945–1953. Objectively speaking, Butler was a creative scientist. He was recognized in physical chemistry as a leading figure within England, and in fact was rewarded with a Fellowship of the Royal Society, a rare gift, before the end of his career.

To picture the scene in which this story takes place, one must think of an office with two tables, one of which was occupied by Butler and the other by Conway, my colleague and friend. Butler had a belief in theory rather than practice, and he would spend many hours at his desk drawing diagrams, making mini-calculations with handheld calculators, etc., before he went to the laboratory to try something in practice. One of the side-effects of Butler’s most intense thinking was whistling. He would do this entirely unconsciously and quite tonelessly. Now this would happen every day, and Conway knew that those times of whistling were particularly times in which Butler could not be disturbed. But the curious part is that, after a

strenuous bout of whistling, Butler would often turn around in a petulant manner and demand, “stop that whistling Conway!” He never understood that he got the wrong person.

Butler story 2

This account describes an event that occurred after Brian Conway had come to work with me at the University of Pennsylvania, where I was in my second year. When he returned to his Institute in London after a year’s absence, he was coming down the stairs when he met Butler, his boss, coming in the opposite direction. Butler wore those gold rimmed, small glasses, and did not see very well, but as the two came near to each other, Butler said: “Oh, err, is that you Conway?” “Have you been away? I have been looking for you.”

Henry Eyring story

The chairman of the committee which was going to decide the Nobel Prize was present at the banquet, and he could not understand why it was that he had to wait so long for Henry, who was nearby at the Academy’s hotel. Finally, the chairman decided to send a limousine to get him to come to dinner, and eventually he did turn up, but how was he dressed? His reputation had always been that he was a great simplifier and he would give lectures on the most advanced quantum mechanics in which he simplified the whole thing down to sheep jumping over fences (or going through them). Despite this mathematical sophistication, Henry had no idea of how Swedes dressed for a formal banquet. It required not only black pants, but tails behind the pants towards the floor. Unfortunately, Henry was dressed in an open neck shirt, yellow, and, horror of all, football shorts. The Swedes are very well mannered people, and of course did not say anything, but that was the end of the Nobel Prize for Henry Eyring.

In 1964, Amulya K.N. Reddy joined our community. He was a post-doc, a senior man, and had been educated in the West although he came from India, indeed at the same Imperial College where I got my own Ph.D. degree. He had accepted an invitation to join my electrochemistry group on a rather tentative basis because he had no detailed knowledge of electrochemistry at all when he joined us, but he was very intelligent and later on we wrote a two-volume book together, and a famous one at that, “Modern Electrochemistry”. In many ways, his lack of detailed electrochemical knowledge was actually a help, because I had to explain everything in detail, and when “the penny dropped” his surprise and joy came through in his good, clear writing.

When I first met Reddy, I had just returned from a trip to Norway, where in the far reaches of the north there is a small

town called Trondheim, famous for a battle between the British navy and the Germans forces in 1940. The man I had gone to see in Trondheim was called Winterbotham, and he possessed an apparatus which was new to me and to others. It was called an “ellipsometer”, and instead of measuring the strength of light beams as they interacted with an object, this apparatus measured the complex reflectance ratio. Although there were many mathematical disadvantages to the technique, nevertheless you could do things with it that were not possible with conventional microscopes.

Thus, when Reddy joined my group, I told him about my visit to Trondheim and the ellipsometer. He grabbed the idea of something new (in which I myself had no experience) and started to work on it, including the mathematics. Reddy proved remarkable at once, because within a few days he had not only mastered the way the ellipsometer worked, but he had also mastered the mathematics. This had not yet been computerized and one had to do every calculation by hand. Soon Reddy set about writing a computer program that would simplify the long and tedious mathematics that were needed. Today, it is possible to purchase a Fourier transform ellipsometer that performs the necessary calculations in seconds, but in Reddy’s time it took more than 8 h to get a single result.

One could measure very thin films on metals down to something like 5 Å, and could also observe organic compounds at less than monolayer coverage of electrode surfaces. This set Reddy up as one of the world’s leading experts in ellipsometry. Looking back and putting into a context so much that was done in the 1950s and 1960s, I would claim that Reddy’s ellipsometer was the beginning of optical methods for looking at surfaces on an atomic level.

Another person who came from overseas was Alexander Damjanović. We called him Sasha. Sasha was a great man for optical microscopy and he knew a lot about new techniques, in particular Nomarski Interference Contrast microscopy. With the latter we were able to study many interesting features of crystals on surfaces, down to a level of about 500 Å. It enabled us to watch the crystals growing in solution, whereas all the other methods (except ellipsometry) required a vacuum and therefore required removal of the sample from the growth environment.

Another technique which Damjanović introduced to us was the rotating ring-disk electrode. Rotating electrodes had been much used before Damjanović’s time, particularly by Russian workers, and Damjanović had some experience of this from a lab in Moscow. By adding a ring to the rotating disk (quite electrically unconnected with it) he was able to show that he could analyze intermediate compounds which existed after the first electron transfer. This was particularly valuable to us in the study of oxygen reduction in fuel cells, and later provided the basis for a more detailed study of the oxygen reduction reaction.

I already mentioned Wolfgang Mehl, who introduced transients into our group at quite an early stage, but there was another man from Germany who was much older. That was W. Beck, and he would always emphasize to us, when he paid us a weekly visit, that he had worked with the famous German electrochemist (and Nobel prize winner) Walther Hermann Nernst. What Beck did for us was to put before us the great need for work on mechanical *stress*. It turns out that in metal dissolution and deposition (the all important problems of corrosion) there are unseen stresses in metals which are often the sites of the corrosion reactions which take place at a later time. Beck was keen on finding out how you could identify these areas of stress, and thus determine how the velocities of various reactions, in particular the hydrogen dissolution reaction, depended upon stress, strain and potential.

Beck's work with us lasted for only about 6 months and he never visited us more than once per week. But one of the things I remember most from my time at the University of Pennsylvania were the results we obtained from the Beck inspired experiments.

This brings me then to speak about Shyam D. Argade, a Ph.D. student who contributed two kinds of things to our program, one of which was fundamental. I am speaking about the calculation of the absolute electrode potential. You would have to be a professional electrochemist to realize that when electrochemists speak about "potential this" and "potential that" they are not really referring to the potential as everybody thinks of it, between the metal and the solution, but to a far more complex quantity which involves also the contact between the material of the electrode and the material of the reference electrode which is measuring it. Argade took this situation to pieces, figuratively speaking, and deduced that the absolute potential of the standard hydrogen electrode was approximately 4.6 V at 25 °C. This result was close to the modern value of 4.44 V.

Among the various interesting characters who joined me at the University of Pennsylvania, the last one I would like to mention is Boris D. Cahan. I used to consult for a company called "Philco" and on one of my visits I came across Boris, who was 33, working hard, at a very much reduced salary compared with Ph.D. graduates, and yet doing the same work they did. I pointed out this discrepancy to Boris, and mentioned that he could increase his salary by 30% if only he would stop for a few years and get a Ph.D. He accepted my invitation and came to work for us in 1963. He was much more than a student. In fact, when Boris was working, you often wondered who was the supervisor and who was the student; and oftentimes he knew more than anyone else in the group. His specialty was new equipment and new techniques, and he began working, in particular, on a technique called Mössbauer spectroscopy.

We had never done anything of that kind before, and I personally knew nothing about it, but this did not deter Boris, who immediately took it up. It was a newly invented technique, with very little literature, but Boris soon mastered it. It had the drawback that it was not applicable to all metals, but it did apply to iron, which of course was super important to us in our corrosion studies.

An electrochemical life

Part 3

Now, let's come to the big break. By 1972, I had been at the University of Pennsylvania, for 19.5 years, when I decided it might be a good idea to continue my work elsewhere. I had been travelling a great deal at this time, in various parts of the world, and made three visits to Australia. I very much liked the atmosphere. Remember I was brought up under British rules and British atmosphere in South Africa, and it all came back to me in Australia. Although a very modern and independent country, I realized that Australia still retained a strong UK connection.

By contrast, there were things which developed at the University of Pennsylvania that I disapproved of. One was the "student revolution". The administrator's attitude was, *let the students win*. They invited them to enter the University without qualifying examinations, and although one could do this in English classes, it was not something to be done in the sciences. Another thing that hit me was the attitude of the new Head of Department, who had very extreme ideas about physical chemistry. He told me that "to do quantum mechanics with something as complex as a metal lined with water was far too difficult". His idea was to make every system simple. Go to low temperatures, use individual particles, etc. He told me outright that he did not think that electrochemistry was a good subject for the Department which he was now about to run. He never told me that I should leave, but it was quite clear that he did not want me.

I had friends in Australia, and one in particular (Harry Bloom) was a Professor in Tasmania. I mentioned to him that I would not mind spending a few years in Australia, and I suspect that it was that conversation which gave rise to a series of phone calls from an individual who kept on telling me that a new University, called Flinders, (after Captain Matthew Flinders RN, the discoverer of South Australia) was looking for someone to run the physical science section of the University. Omitting details, I joined Flinders University in 1972.

Two people at Flinders stand out in my memory. One was a man called R. Winston Revie. I never met anyone who showed more enthusiasm for his work than Winston.

He had a Ph.D. from the Massachusetts Institute of Technology. The other person who turned out to have world recognition in his field 10 years after he left me was a Japanese scientist called Kohei Uosaki. He was one of those super-energetic people who would run between locations. He was known among the other students as “the locomotive” and he worked though his Ph.D. in a short 2 years. His topic was photoelectrochemistry, i.e. the relationship between light directed at electrodes in solution, and the resulting decomposition of water.

For a brief period I think that Exxon and Co. were genuinely worried about research on photoelectrochemistry, because if it had been acceptable cost-wise, the entire petroleum industry might have been threatened. Indeed, I had a number of indications (which I shall not go into here) that I was not out of sight of the CIA and their long arm whilst down in Australia. They always knew what was going on in energy research.

Uosaki had several good ideas whilst down in Australia. One of them was to illuminate both electrodes in a photo cell instead of just the one that other people had been using. Not only can one hope for a doubling of yield by doing this, but also the really fruitful thing that came to light (fully developed later, back in Texas) was that if you sprinkled a small amount of catalyst on the electrode, and was careful not to exceed a monolayer, then you could obtain a tremendous increase in the efficiency of conversion of light to hydrogen. I will say more about this later on.

I must not forget Harvey J. Flitt, who worked very hard to develop a means of detecting stress corrosion cracking. Harvey’s successful development of a laser-based method for picking up tiny amounts of hydrogen in metals was important, though he did not finish his work in thesis writing till I returned to America.

Back to America (1978)

In 1978, I was appointed Distinguished Professor of Chemistry at the Texas A&M University, College Station, Texas, USA. Back in America I wanted to go on a new pathway in my research and I had promised many people for years that I would go into biology and see what aspects therein might succumb to modern electrochemistry. Years ago, I published an article with Supramaniam Srinivasan in which we drew attention to the fact that the metabolism of living creatures was remarkably efficient, i.e. about half the energy we take in as food actually gets converted to energy. Looked at from a non-biological point of view, this is a very high percentage, and it is very difficult to explain unless there is some kind of “fuel cell substitute” working in the body. Shortly after arriving in Texas, I encountered a lady called Agnes Rejou-Michel, and together with M. Ahsan Habib we carried out some experiments to establish

that electron transfer could occur at biosurfaces in contact with ionic solutions.

We also had some limited success in getting electrical power out of bio-fuel cells based on synthetic lipid membranes formed on metal substrates, but these ideas were not readily accepted and developed by others, at least not immediately. Were I to be able to work in the lab right now, then that is a subject that I would certainly like to develop.

A negative experience for me during the early years back in the USA was the controversy about solar energy, and the extent to which it might challenge conventional sources of energy. Eventually the politics became very distracting. Fortunately, this was partly offset by the arrival of another new colleague, Vladimir Jovancicevic. He was a tall learned-looking person who was accompanied by a most interesting wife, who made a living from paintings which were of stalinistic art. Jovancicevic was a great boon to my work as he got me back away from the sensations of the conversion of light to electricity and squarely into normal, but advanced, research chemistry. He did two main things whilst with me. Several people had examined reduction of oxygen which comes into every fuel cell, and is the one bit of fuel cell work still open to academic research. But the systems they had chosen were always at the extremes of pH, namely acid solution and alkaline solution. No one had worked in the middle, in neutral solutions.

Jovancicevic made a good job of deducing the mechanism of oxygen reduction in neutral solution. His particular apparatus was one which we had developed earlier, the rotating disc with ring. This method allows you to find the intermediates of reactions, and if you know these and how they behave, you are well on the way to understanding the rate determining step of an electrode reaction.

Another thing that Jovancicevic did was that he showed how the ellipsometer could be used as a spectrometer, i.e. one could vary the wavelengths of the light which were falling on electrodes and subject them to ellipsometric analysis. This did not take Jovancicevic very long, maybe 6 months, and it changed ellipsometry into a really viable tool.

Before I get back to more technical matters, I would like to mention that we were not averse to a bit of sociology. Balasubramaniam Dandapani was a person who had worked in American industry for 20 years before he decided to come and give us the benefit of his knowledge. But, he did not make fast progress on any electrochemical topic I gave him, so I switched him to examining something I thought might be of more general interest. “What was the relationship” I asked, “between the amount of energy nations put into their populations and the living standard it produces?”

Of course this meant about 3 months research in the library, but eventually Dandapani turned up with a graph which was full of information. On the ordinate, or upward part, he printed the average salary per head of the

population, for about 30 nations. Along the abscissa, or horizontal part, was the average energy, also per head of the population (the energy including everything to do with the military). The shape of Dandapani's curve was, understandingly enough, for low energies, perhaps 2 kW per person, and the living standard was low, i.e. people were on the starvation level. But as the amount of energy was increased, there was an exponential rise in living standard, which rose towards energies like 5 and 6 kW. Then it bent over, and became flat, and there was no more increase in the living standard by feeding more energy into the country. This seemed to me to have significance. At that time (1988), the USA was the world's largest consumer of energy at 10 kW per person. But, as I knew from my life in America, it was also an easy-going, wasteful, society in which many families had three or four cars. Therefore, it seemed to me that if one cut back the amount of energy that was used by an advanced nation, its living standard would hardly change. This idea has come into all the work that I have done more recently (indeed after retiring from the University) about the supply of energy which would be clean and inexhaustible.

A long-term colleague of mine at the University of Pennsylvania was Alan MacDiarmid, a very energetic New Zealander who came to Penn about the same time as I. Alan had previously received a Shell graduate scholarship to study silicon hydrides at Cambridge University, England under the directorship of H.J. Emeléus. After a brief appointment as a junior faculty member at Queens College of the University of St. Andrews, Scotland, he accepted a junior position on the faculty of the Department of Chemistry at the University of Pennsylvania. Alan, together with Alan J. Heeger, and Hideki Shirakawa, won the 2000 Nobel Prize in Chemistry "for the discovery and development of conductive polymers".

MacDiarmid, Heeger and Shirakawa made their seminal findings at the end of the 1970s and conductive polymers quickly developed into a research field of great importance for chemists as well as physicists. Their discovery seemed to hold immense promise and I fell upon it from the electrochemical point of view. I reckoned that it would be easy, using electrochemistry, to make fairly slight changes in the surface of these electronically conducting polymers so that they would act as photo-electrodes, and by varying conditions appropriately, I could make a reaction go faster or slower. I still have grand fantasies of being able to patent a fuel cell electrode which can be easily and cheaply made from polymers rather than metals.

Cold fusion

It is now 1989 at the Texas A&M, and in order to account for the atmosphere and happenings, I have to remind you

that when I was at Imperial, I knew a student who was only 2 or 3 years behind me in his Ph.D. (and also in his age). His name was Martin Fleischmann and he appeared on *The MacNeil/Lehrer NewsHour* (a broadcast TV program) in March 1989, together with a colleague named Bobby Stanley Pons, to announce the discovery of something which they thought would cause a great sensation (as it did).

What they claimed was that, by chemical means, they had made a reaction occur which was *nuclear* in nature and not chemical. Of course everyone knows that nuclear reactions can cause bombs of incredible strength, a million times more powerful than ordinary bombs. So everyone expected a nuclear reaction to be much more powerful than a chemical reaction. And, so, at the time of the announcement, every scientist was up in arms at the ridiculous nature of their claim. It was also widely noted that they were chemists not physicists.

All the nuclear physicists proclaimed, as one man, that the idea could not be true. But the chief experimental result that Martin Fleischman used to back up his claim was the release of heat. He knew the amount of heat that a normal electrolysis cell should put out (it is not ambiguous), and he found that his cells were sometimes putting out more heat than that. Now I must make one thing clear. It disturbs the truth to say that he was "electrolyzing water". What he was actually electrolyzing was D₂O, deuterium oxide. He used deuterium oxide and evolved *deuterium* and *oxygen* instead of hydrogen and oxygen. It was this kind of electrolysis which he claimed was releasing additional heat. Within days the world had divided itself into two parts. 99% scoffed at the idea, saying it must be a mistake, and 1% thought that, hey, maybe it is wrong, but suppose it is right...?

Now, I would kept a loose connection with Martin over the years, seeing him at scientific conferences, etc., but it would be untrue to say he was a personal friend of mine (which some made out). When I saw the controversy shaping up after the TV appearance, I knew what I had to do; I had to see if he was right. At the time, I had 23 coworkers, a large number. I took 19 of them and asked them to work for a few months on proving or disproving the Fleischmann–Pons hypothesis. However, as a true scientist, I chose to look at it differently from the way Fleischmann did. I did not seek to repeat his excess heat experiments, because the heat that he was measuring was not great, and errors in measuring heat are easy to make. For example, at first we did not realize how important it was to stir the solutions to be sure the temperatures were uniform, and many other things that were indeed looked at by others who tried to replicate the Fleischmann–Pons results. I went a different route. I knew that deuterium was the second isotope of hydrogen, so I decided to look for the

appearance of the third isotope, called tritium. What egged me on was that tritium was radioactive, so I saw a wonderfully quick experiment.

This story goes on, so I have to cut it down. One of my students was called Nigel Packham. At the beginning, we had no special instruments in the chemistry department to give us a signal, and so we relied on the applied nuclear engineering department. It was Nigel who first came back from a visit to them, and, referring to one particular sample, said simply, "... he said this one is full of tritium". He was referring to a technician who had analyzed for tritium. It was his fourth visit and all the other cells had shown no tritium. Well, of course it seemed sensational, but we could not publish a paper on the basis of one result, so we had to wait a few weeks to see what other students got. It was not all that certain. Yes, there was tritium. No, there was no tritium. After some weeks it appeared to us that one cell in four gave tritium and it seemed worth reporting, but it was not that certain.

Now the rest of the story is not too nice, because when we said we had tritium, you could say the world imploded. The response was vastly negative. The controversy was all over the NY Times and other journals, and whatever we said in support of Fleischmann was immediately cancelled by various negative persons (particularly two on the West Coast) who kept on attacking us, and who said it could not be done, we had made errors, etc. Well, to make the story super-short, the saga went on for 2 or 3 years. Of course I continued my research and got tritium a total of 18 times. But, the weak part was, I could not promise a specific day on which to come and see a positive result, because I did not know if tritium was being formed on that particular day.

Then came a new post-doc, Chun-Ching Chien, from Taiwan. I was assured by the late Norman Hackerman that he had seen Chien make tritium on a visit to Thailand. (Norman was a Texan, very tough, and he had been the chairman from 1974 to 1980 of the National Science Board, which is composed of 25 members appointed by the President and confirmed by the US Senate, representing the broad US science and engineering community.) I promptly appointed Chien and he came and worked with me for a year. Well of course I did not only want to find tritium, I wanted to find something else: because when we took Fleischmann's heat and compared it with what the production of tritium would give, the numbers did not add up. Was the heat real? There must be something else, and everyone said: *helium*. I knew, as a scientist, that they were talking about $D+D \rightarrow He$, an obvious alternative if one electrolyzes deuterium oxide. So I set Chien a task: "Find helium!" Well it was not so easy, for helium is an inert gas and does not react with anything chemical. But it can be detected with a super-sensitive mass spectrograph. Initially, we thought we had detected He^3 rather than the He^4 that we

were looking for, but after a couple of days we were able to find an electrode producing both He^4 and tritium. So the heat was real, and Fleischmann was right.

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