‘Rutherford’s Chemists’

Special Issue

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Contents

Introduction 3

Frederick Soddy: Transmutation in Science and Society
  by Linda Marie Richards 6

Marie and Pierre Curie and their Discoveries
  by Pierre Radvanyi 17

Radioactive contamination of the laboratory notebooks of Sir William Ramsay and Frederick Soddy: historical and radio-archaeological perspectives
  by Neil Todd 20

The early influence of Manchester on the work of George de Hevesy’
  by Siegfried Niese 57
Rutherford’s Chemists

This is our second ‘online only’ issue to be published following ‘The History of Particle Colliders’. As with that meeting it was never intended to produce a written record, but once again we are very fortunate that some of the speakers have been able to write up their talks and I have great pleasure to present these as a special online issue.

My thanks to Pierre Radvanyi, Linda Richards, Siegfried Niese, Neil Todd,

Malcolm Cooper
Editor

This was a two-day meeting, co-sponsored by the Institute of Physics and Royal Society of Chemistry, to celebrate the centenary of the second scientific revolution and a unique collaboration between physics and chemistry. , 15th/16th July, 2017

Welcome from the Department of Physics, University of Glasgow

I would like to welcome the Institute of Physics, History of Physics Group, and the Royal Society of Chemistry to Glasgow to this meeting on Rutherford’s Chemists. This meeting has been organised largely through the enthusiasm and efforts of Neil Todd. I will start with a brief note about our own history.

Our most famous Physicist was William Thomson, later known as Lord Kelvin. Thomson was appointed to the Chair of Natural Philosophy at age 23, in 1846. He retired some 53 years later in 1899, before going on to re-matriculate as a research student. He was active in physics till his death in 1907. Today’s meeting is actually being held in the Kelvin lecture theatre. Modern nuclear physics was established in the Department of Natural Philosophy following the appointment of Philip Dee to the Chair of Natural Philosophy in 1943. The first experimental facility was a 330 MeV electron Synchrotron commissioned in the basement of this building in 1954. Staff subsequently split into experimental High Energy Physics and Nuclear Physics Research Groups. Later theoretical Nuclear and Particle Physics groups were also established. It is very appropriate that this Rutherford’s Chemists meeting is being held in Glasgow. The first Scottish connection is that Rutherford’s father, James, was a farmer from Perth who emigrated to
New Zealand, where Rutherford was born in 1871. The earliest Nuclear Physics work was actually carried out in Chemistry departments, as radioactive properties were first associated with particular elements. Indeed, Rutherford obtained the 1908 Nobel Prize for Chemistry in recognition of his work into the disintegration of the elements and the chemistry of radioactive substances. It is only following the work of Rutherford and Bohr, and those that worked with them, that we now appreciate that radioactivity is actually associated with the physics of the nucleus. Some sixty years after his death in 1937, the element Rutherfordium, with an atomic number of 104, was named in Rutherford’s honour. A second Scottish connection, which we will hear a lot about during this meeting, was the work of Frederik Soddy, one of Rutherford’s Chemists, who worked in our Chemistry department. He was closely associated with developing the concept of isotopes.

*Douglas McGregor*

**Welcome from the Organiser**

May I also add my welcome to this meeting “Rutherford’s Chemists”. Its genesis has multiple sources, one being a meeting entitled “Rutherford’s Geophysicists” which was held in Cambridge a few years ago and which was considered to be very successful, so providing a good model. Another was that about the same time in 2013 the centenary occurred of some very important landmark discoveries, including the birth of the quantum atom, the isotope, atomic number which allowed the formulation of the modern period table. These discoveries had followed on from the discovery of X-rays, radioactivity and the electron which had triggered a period of dramatic change in physical science, referred to by some historians as the “second scientific revolution”. It was a unique period in the history of science which one could argue has never been matched before or since. One person stands out from this period of science, none other than the great Ernest Rutherford. Unravelling the mysteries of radioactivity, though, was a task which required a multi-disciplinary approach, not least one which drew heavily on the disciplines of both physics and chemistry, hence the particular theme of this meeting. Rutherford, himself, famously was awarded the Nobel Prize for Chemistry.

The Institute of Physics and Royal Society of Chemistry History Groups collaborated most recently during the centenary of HJG Moseley’s work on the high-frequency X-ray spectra of the elements. And it was this joint
effort which gave the final impetus to make the present meeting happen. The question ‘where should the meeting be held?’ was in part determined by which radio-chemists we should choose to have represented at the meeting. Among that band of scientists, again, an outstanding figure was that of Frederick Soddy. Soddy worked in a number of locations, after the seminal collaboration with Rutherford at McGill, (for which it has been argued that he should have been jointly awarded the Nobel Prize with Rutherford) but the work for which he *was* awarded the Nobel prize work was carried out here in Glasgow.

I had another reason to select Glasgow; for some years previously I had been in contact with Professor David Sanderson concerning some historic radio-active artefacts associated with Soddy. David contacted me given my interest in the history of radium because at that time the UoG was considering disposing of the artefacts. Thankfully, these are preserved and are being kept in the Hunterian Museum.

Having selected the city of Glasgow I was happy to make contact again with David and I should express our thanks to him and to the other members of the Department of Physics here, not least Professor Martin Hendry.

It is right that Soddy is represented at this meeting, but there were, of course, many other chemists who made important contributions, and these are acknowledged in his various books, not least the Chemistry of the Radio-Elements Part II, which was written after the concept of the isotope and the displacement laws were fully articulated in 1913. In the early days of discovery, manufacture and distribution, we might mention the work of Friedrich Giesel and Stefan Meyer, and in the early days of interpretation at McGill with Rutherford, Harriet Brooks with the Curies, and André-Louis Debierne. In the contributions leading up to the final formulation of the isotope, major contributions were made by van den Broek, AS Russell, Georg von Hevesy and Kasmir Fajans, and in later work, such as the interpretation of nuclear fission, Lisa Meitner. I think we’d really need several days meeting to do justice to these and others, but given time and other limits the current set of talks I feel gives a reasonable coverage of some of the major workers. Finally, then, I should offer thanks to the speakers who have made time and effort to be here today.

*Neil Todd*

*University of Exeter, Exeter, UK.*
Frederick Soddy: Transmutation in Science and Society

Linda Marie Richards

Oregon State University, Corvallis, USA.

It is fitting Sir Ernest Rutherford and Frederick Soddy (1877-1956) first developed their relationship in a public debate over atomic matter. From then on, Soddy was consumed by what mattered most about atomic energy, for good and ill. Soddy embodied a rare duty to explain radioactivity’s relationship to time, matter and the very meaning of human existence, as the first person to comprehend the meaning of atomic forces. This obligation directed Soddy’s life. He came to see nuclear weapons as proof of the need for scientists and civil society to take more of an active role in their own destiny, beyond “false profits” and war.

Soddy’s actual chemistry and his concerns of how nuclear science and technology in general might transmute society can inform us today. This summary focuses on just three aspects of Soddy’s complex contributions. Soddy’s collaboration with Rutherford offers the benefits and risks of interdisciplinary work, while Soddy speaks to the dangers of science, war and nuclear contamination. Soddy’s best intentions offer a tragic culmination of his once beloved radiation.

Soddy and Sir Ernest Rutherford at McGill University

The initial discovery of radiation in the late 1890s caused a sensation---but it defied explanation. It was not until the work of Soddy and Rutherford that a consensus emerged on how radiation worked. Some of Soddy’s collaborative research with Rutherford while at McGill University contributed to Rutherford’s 1908 Nobel Prize in Chemistry, despite Rutherford being a physicist. This boundary crossing is important today when problems are equally as complex and may be best approached by similar interdisciplinary efforts, despite the tension and losses.

When Soddy first came to McGill in Montreal to teach in May of 1900, he felt strongly as any trained Oxford chemist that atoms were not divisible. This mistaken but vehement belief on his part is what led to him meet Rutherford. The two men first met when they opposed each other in a public debate hosted by the McGill Physical Society, a group chaired by Rutherford. The debate intended to showcase Rutherford’s recent research into thorium emanation and what he called alpha and beta and gamma radiation.
The debate was not as easy as Rutherford hoped. He was surprised by Soddy’s scathing dismissal of physicists’ work, many by name. Soddy was relentless. For example, he ended the first debate with this salvo: “Possibly Professor Rutherford may be able to convince us that matter as known to him is really matter as known to us, or possibly he may admit that the world in which he deals is a new world demanding a chemistry and physics of its own, and in either case, I feel sure chemists will retain a belief and a reverence for atoms as concrete and permanent identities, if not immutable, certainly not yet transmuted.”¹ His words, meant to discredit, actually predicted how Soddy later described radioactivity.

The discussion simmered and extended into the next week. Soddy’s articulate wit and scientific precision, if not his belligerence, impressed Rutherford. Soddy was equally struck by Rutherford’s investigations into the problem of radioactive behavior. Considering Rutherford’s ongoing research and J.J. Thomson’s indisputable evidence of electrons, Soddy inverted his original debate position. Within a few months of their combative first debate, Rutherford invited Soddy to join him. Rutherford needed just such a traditionally trained chemist as Soddy; Soddy having been a gifted student at Merton College was also experienced as an independent researcher at Oxford for two years before becoming a demonstrator at McGill.

The scientists coauthored nine papers in nineteen months. Their correspondence shows the warmth Soddy had for Rutherford, and his early attempts to diplomatically navigate his position as a young non-tenured chemist versus Rutherford’s prestige. Soddy knew, however, the value of comingling their disparate ideas and often asks for Rutherford’s point of view. For example, in a July 24, 1902 letter Soddy tells Rutherford his conception of how thorium gas can be measured and asks Rutherford to share how he pictures it. In the same letter he writes,

“You are quite right that I should get myself in trim for working with the devil, I mean like the devil with you. Some of the work can certainly be much better done in partnership”².

² Rutherford-Soddy Correspondence, Courtesy of American Institute of Physics.
By 1902 Soddy and Rutherford determined radioactive gases were being emanated from what was originally an inert sample of thorium. The thorium had somehow become, with no matter lost, and nothing altered, some new intermediate substance expelling radioactive gas. Soddy intuited (perhaps due to his fascination with alchemy) they were seeing something like the transmutation of elements evoked by alchemical lore, an interest of his. Atoms of thorium, he surmised, autonomously transformed into some other radioactive substance. Ironically it would then be Soddy who needed to convince Rutherford of the revolutionary process Soddy coined “transmutation” was indeed real.

This example of the men suggests how rich alliances between opposite poles might be, considering Soddy’s position against such an interpretation at their earlier Physical Society debate. Rutherford and Soddy blended their individual expertise in physics and chemistry to the problem, each considering it very differently. This advanced them beyond other research groups already working on the problem who were siloed into one way of seeing in their own discipline. That the two opposite points of view converge in the two men speaks to the beauty of accepting different points of view in our time of polarity and the alchemical inspiration speaks to the contribution of imagination and liberal arts to science. What the men saw, albeit each in their own laboratories, could only be explained by what we they created together, the disintegration theory of radioactivity.

But the tensions between the men are instructive, too. Soddy never did entirely agree with Rutherford and vice versa on many fronts. Soddy defended the atom as what gave elements their identity and integrity, despite their divisibility on the atomic level. Soddy reconciled these conflicting versions of atoms in his public writing with what he called a chemical atom, deftly integrating their find into the laws of traditional Newtonian physics.

In their correspondence, you can see the stress of competition. Soddy was invited to publish a summary of their work for *The Electrician*, which became his 1904 book *Radio-activity*. Rutherford was simultaneously publishing his own writing on their work together. Soddy wrote to Rutherford defensively that he meant no harm but in sum, if Soddy did not write up the chemical side of transmutation for *The Electrician*, someone else less informed about their work together would.
Soddy was the more worldly, philosophical writer with a much wider vista than Rutherford. Soddy, not Rutherford, initially grabbed the public imagination. Soddy framed the meaning of their find steeped in evocative alchemical terms, immediately amplified by an exuberant press. Soddy was much more confident than the more careful Rutherford about the value of their work. Soddy stated as early as 1903 that the process of transmutation could be harnessed to create energy and for miracle cures like the philosopher’s stone for universal health. Soddy felt atomic cures and energy held the potential to create abundance, health and equality; a transmutation of the social order.

Soddy embodied the leap science had just taken. He wrote to Rutherford on August 7, 1903 “Having failed utterly as I can see to make you realize the width of the gap between our recent and anything preceding, I do not intend to attempt that it in this letter so you must still continue to smile as the thought of the library discussion of more than two years ago comes back to you” perhaps alluding to their initial debate and Soddy’s harsh rejection of any type of transmutation being possible.

The risks of losing identity in a collaboration are also clear. Soddy suspected he was invisible in their collaborative work. He wrote to Rutherford “…heard some naive remarks that you must be a very good chemist as well as a physicist” (March 7, 1903). Soddy also wrote about his allegiance to his discipline, “I sometimes feel however, I had been traitor to my own camp and let (illegible initials) the enemy in by the backdoor.”

In Soddy’s letters to Rutherford, their correspondence shows affection and Soddy’s respect for Rutherford. However, Rutherford with his more intimate friend Bertram Boltwood shared impatience, anger and biting dismay at Soddy’s seemingly stubborn attachments, perceived slights and indignations. For just one vivid example, in 1909 Rutherford wrote, “He can never state his results without hitting someone else…Soddy reminds me of a cock crowing on his own dunghill.”

Soddy’s work with Rutherford can best be located in between chemistry and physics, in the flexible field of physical chemistry. Soddy did not approve of this lineage however: to add the word “physical” mistakenly truncated chemistry, limiting its breadth inaccurately. Soddy later in life, bitterly complained about the usurpation of radiochemistry by the term and dominance of nuclear physics. Soddy felt his own theory of disintegration proceeded Einstein’s theory of relativity. He doubted much about physics,
from the confirmation of the theory of relativity by Eddington to the logic of mathematical probabilities relating to the infinite. In short, he was infuriating to many physicists.

The future, along with Soddy’s other passionate choices, exacted a price for this. In fact, in a 1951 BBC interview, Soddy said he had become “dead to science”. In later parlance, the radioactive decay coined by Soddy as transmutation would be called instead transformation, which refers to matter changing on its own, just something observed, passive and eviscerated of vitality. It would be Rutherford, not Soddy, who achieved the true feat of transmutation, the manipulation of matter, in 1919.

**Soddy and Nuclear Weapons**

Soddy as a youth was concerned with the distribution of wealth. He saw science and technology as a way to improve human lives on a practical scale by ending poverty and war. By the time Soddy was in his mid-twenties, he had left McGill for London and confirmed the theory of disintegration with Sir William Ramsey. Soddy felt selected to translate radioactivity into social change. For artist Salvador Dali, nuclear weapons proved God existed, an exaltation he captured in his painting “Christ of Saint John of the Cross” (1951). For Soddy, the exaltation he said he experienced having been given a glimpse of how the atomic and chemical structure of the universe was determined proved not God, but his own responsibility as a very young man to transmute the social order.

Soddy believed the discovery of radiation was so profound it completely destabilized the meaning of relationships, time and matter. As he wrote in his 1909 book *The Interpretation of Radium* “Today while all is strange and new, and the very name of the science even unfamiliar, it may appear to be a far cry to attempt to foretell the effects these discoveries, concerned primarily with the ultimate nature of matter, are destined to exert on our conceptions of the ultimate destiny of man” (7).

While most of Soddy’s early popularization of radioactivity held out promise for healing, abundance and equality, he had no doubt about the negative potential power his researches might unleash. Lecturing the Royal Engineers in 1904, Soddy said:

"The man who put his hand on the lever by which a parsimonious nature regulates so jealously the output of this store of energy would possess a weapon by which he could destroy the earth if he chose."
In 1904, Soddy was hired by chemistry chair and alchemical historian John Ferguson at the University of Glasgow as the first lecturer in Physical Chemistry and Radio-activity. Glasgow, with its radical environment fighting for women’s equality, labor rights, and support for science, was the perfect environment for Soddy to consider his ideas about how transmutation might alter society for good or ill. He was also thinking in terms of the actual energy and the economy.

Soddy recognized a relationship between access to energy, poverty and war. He anticipated ecological economics with his book 1912 book, Matter and Energy. Both his radiochemistry and his work for social improvement through science however, were soon interrupted by war. He was chair of Chemistry at Aberdeen during WW I when practically all research became subsumed to military ends. By 1914, Soddy cooperated only to work on defensive responses to chemical gases. His opposition to war was only deepened by the loss of brilliant Oxford chemist Henry Moseley, who died a pointless death as a telecommunications officer at the Battle of Gallipoli April 10, 1915. Moseley’s death made Soddy furious. The use of chemical weapons, developed out of the agricultural endeavor to feed the world through nitrogen fixing, convinced him beyond a doubt science would only increasingly serve as a tool of war and false profit.

Soddy’s predictive imagination inspired HG Wells to write The World Set Free, a fiction novel published in 1914, suggesting how weapons could be made from atomic discoveries. Wells’s book included the first, albeit fictional representation of a chain reaction, based on Soddy’s earlier musings in The Interpretation of Radium. Leo Szilárd read The World Set Free in the 1930s and conceived of how a neutron chain reaction could work. This led to a reversal of Soddy’s best intentions: to nuclear bombs, which Soddy feared, and later, nuclear power, which Soddy thought could create equality by distributing access to energy fairly in an unjust world. Disturbingly Wells’ fictional "atomic bombs" induced "a blazing continual explosion" that it is "never entirely exhausted," evoking Chernobyl and later Fukushima as gaping eternal radiant wounds.

What mattered most to Soddy about atomic matter went far beyond chemistry to direct his trajectory into economics. Soddy abandoned much of his radiochemical research by 1919 and turned to the invisible underpinnings of social structure, in the hopes of intervening in the economy. He observed unlimited debt was available for waging war without funds to improve the human condition. Soddy’s economic work
predicted the monetary system, based on unlimited debt with perpetual
growth, was dangerously out of alignment with the reality of decreasing
resources and increasing consequences like smog, pollution and ill health.

After the First World War, he protested the relationship of science to
weapons making in articles, books and even his Noble speech. He received
the 1921 Nobel Prize in Chemistry in 1922 for his work on isotopes,
primarily, but in addition—just as Rutherford’s award read—-for his work
understanding the “chemistry of radioactive substances.” Alluding to the
“darker side of science” Soddy pronounced:

“As a scientific man I wish to acknowledge and accept my own share of
responsibility and blame for this. As scientific men we have all, no doubt,
 felt that our work has been put often to base uses, which must lead to
disaster. But what sin is to the moralist and crime to the jurist so to the
scientific man is ignorance. On our plane, knowledge and ignorance are the
immoral adversaries. Scientific men can hardly escape the charge of
ignorance with regard to the precise effect of the impact of modern science
upon the mode of living of the people and upon their civilization. For them,
such a charge is worse than that of crime.”

Soddy’s thinking is credited with sparking self-reflection among scientists
and stimulating the anti-technocracy movement in Europe during the 1930s.
Soddy is not only being thought of now for his economics. Some of his
main arguments against war also tied in to economics and the need for a
shift to conservation and limits to growth. Soddy predates and inspired the
ecological economics developed in the 1960s now known as the de-growth
movement. Like Nicholas Georgescu-Roegen, Herman Daly and Kenneth
Boulding, but years before their work, Soddy had seen energy as the system
of life support on the planet and after it is spent, the entropy and chaos. That
entropy, manifests in externalized consequences of pollution, contamination
and disarray.

Soddy felt true wealth could only be measured in terms of thermodynamics,
energy and its availability. Soddy’s prophetic imagination grappled with the
varied effects of nuclear technology from automation replacing workers to
the dangers of a lack of meaning in a consumer society. From 1919 to 136
he was the chair of chemistry at Oxford, where he after he redesigned the
chemical teaching laboratory for serious research, he retired early, the year
his beloved wife, Winifred, died of cancer.
On August 18, 1945, only nine days after a U.S. plutonium bomb decimated Nagasaki, Soddy published a piece called “Tomic Era” in Cavalcade. He dropped the letter “a” off of “atomic” because the word for atom meant indivisible. He was adamant words ought to be precise to describe the actual atomic bomb was made by a process not about wholeness, but its opposite, actually one of the fissioning of atoms to release energy. His insistence was partly connected to his effort to make radiochemistry understandable to the public. He felt all people would be eventually affected by the power to harness this energy, and everyone should be literate enough to be included in the discussion. In “Tomic Era” subtitled “The Week of Destiny Aug 5-11 in the 45th year of the Tomic Era” Soddy writes with fury:

“Vesuvius, Mt. Pelee and uranium have vomited. The world, sickened of slaughter, vomits.”

He saw the events in Japan as sending the message (which he bolded in the text) there would be “control of the world by the atomic bomb, already a fait accompli”(4).

Much to his despair, and despite his efforts such as his 1949 book The Story of Atomic Energy, Soddy’s opposition to war, along with his beloved chemistry, were edited from what became a laudatory physics narrative. The term nuclear physics came to describe the field Soddy claimed for radiochemistry, excising his own contributions with those of chemistry. There is no escaping that while Soddy initially had free reign at imagining with the public all the possible outcomes of nuclear technology, over the course of his lifetime, Soddy could never again command equal attention to warn as he had once encouraged.

Soddy as Memory

Soddy reached far beyond science to frame transmutation as a new kind of alchemy for mankind. Soddy consistently mapped his ideas about science onto society, like Linus Pauling’s much later “molecular idea of civilization”. In sum, both physical chemists saw function follows form and thus, if human structures and organization were changed, this would alter relationships. The way the two men perceived how the world worked according to chemistry gave them ample evidence humans could end war if the shape of society changed. Hydrogen bombs and thermonuclear weapons presented dire conditions, demanding leadership among scientists to find
equally experimental ways to organize global and human relations without war and violence. Both men signed the Mainau Declaration of 1955 stating,

“All nations must decide voluntarily to refrain from violence as the last means of politics. If they are not prepared to do so, they will cease to exist.”

Soddy died the next year, unable to construct a chemical “tomic” memory for the 20th century. Physics, with its gravitas, cyclotrons, reactors and nuclear explosions, claimed the story of radioactivity. But new interpretations are being written such as Mark S. Morrisson’s detailed treatment of Soddy in his 2007 book Modern Alchemy. Other works by historians of science include Louis Campos’s Radium and The Secret of Life who uses Soddy’s detailed biography as frame for the vitalism in his book; Angela Creager’s 2013 Life Atomic which gives a new perspective using isotopes; Gabrielle Hecht with her focus on uranium mining; Jacob Darwin Hamblin’s latest book, Arming Mother Nature, shows how scientists considered the planet itself a weapon during the Cold War arms race. It is altogether a different story now.

Soddy’s life may hold new resonance. This is partly because radiation exposure converges around the chemistry of contamination. This is one of the essential discussions of our time, from the water of the Lakota tribal people threatened by the Dakota Access Pipeline to the chemistry of pollution at colossal wasted nuclear landscapes or nukescapes such as Hanford, uranium mining sites that extend for as far as the eye can see at Elliott Lake, Canada and the Navajo Nation, and now always in the background of the words “nuclear power”– the debacles that are Fukushima and Chernobyl.

The long lived insipid nature and severity of nuclear pollution lends new importance to the chemistry of contamination. From the historical reconstruction of radiation dose studies to radioactive archeology at contaminated former laboratories and using radioactive notebooks, the coming dominant narrative explores radiation as pollution writ large. This longer-lived narrative contains impacts from the biochemical signature of radioactively contaminated human bodies to the ocean currents of the Pacific.

Soddy, even though he was mesmerized by radiation, serves an important prophetic function thus far overlooked in his “Law of the Conservation of Radioactivity.” As early as 1903, Soddy reiterated the lack of human
control of radiation “a process which lies wholly outside the sphere of known controllable forces, and cannot be created, altered or destroyed.” Time proved “transmutation” made creating radiation quite possible by 1919. As to how to destroy it, there remains no answer. Soddy explains we are powerless to control radiation while at the same time he exudes romanticism: “There is something sublime about its aloofness from and indifference to, its external environment” (37, 1909). Soddy acknowledged his ignorance of the biological effects, writing, “The physiological effects of the radium emanation are imperfectly investigated and are probably potent. This is a field of investigation I personally have no desire to explore, so that we must not forget to cork the globe and so prevent the emanation from diffusing out into the air of the room” (112, 1909). Soddy blamed his exposure to radiation for being unable to conceive children and later, for the early death of his beloved radio-activist researching partner and wife, Winifred.

His ideas, like Linus Pauling’s, transcend time into the current discourse on access to energy and the intergenerational injustice of poverty, toxic pollution and climate change. Soddy had foreseen the end of the supply of fossil fuels, especially coal and oil. He was concerned about entropy and depreciation, but he somehow did not fully appreciate the more tangible legacy of nuclear pollution even as he decided radiation could never be controlled by humanity. In his recent rehabilitation by writers and economists Soddy’s memory inscribes our thinking with the unintended consequences of good intentions and a large dose of sobering responsibility. Solving complex problems like the specter of nuclear war and contamination requires unexpected collaborations. This will create new kinds of risks, but as Soddy found, some leaps of imagination are exacted by fate.

Select additional resource list:


University of Oxford, Bodleian Library, Professor Frederick Soddy Papers. University of Glasgow Archive Services, Soddy Papers and Andrew Kent collection.

The author would like to thank the organizers and participants at the “Rutherford and his Scientists” University of Glasgow workshop, who all informed my thinking on this piece. I am especially indebted to Michael Jewess, John Faithfull, Neil Todd, Malcolm Cooper, and Claire Daniel, the accommodating archivist at the University of Glasgow. American Institute of Physics archivist Audrey Lengel kindly assisted me in obtaining the Rutherford- Soddy correspondence. Thank you to former OSU graduate student Elizabeth Nielsen for her assistance in researching Soddy’s early work on therapeutic uses of radioactivity.
At the very end of 1895, from Würzburg, W.C. Röntgen sent copies of his article on his discovery of X-rays to several well-known physicists in Europe. One of these was Henri Poincaré who announced and commented on it at a session of the Academy of sciences in Paris; he suggested that these rays could be a by-product of phosphorescence. Henri Becquerel (1852-1908), member of the Academy, possessed a very phosphorescent Uranium mineral. Back in his laboratory at the Museum, Becquerel tried and observed another new type of radiation, the « Uranic rays » (March 1896).

In November 1891, Marie Sklodowska Curie (1867-1934) travelled from Warsaw (where women were not allowed to study at the University) to Paris in order to study at the Sorbonne. After successful studies, she worked on the magnetic properties of steel. On this occasion she met Pierre Curie (1859-1906), whom she married in 1895. Pierre was already well known for his work on piezoelectricity, on symmetry and on magnetism. However Marie and Pierre worked together on a completely equal footing. Their first daughter, Irène, was born in 1897. Marie wished to prepare a PhD. - she would be the first woman in science, in Paris to achieve such a goal. She decided to search for other chemical elements possibly emitting the same kind of rays as uranium. She used a quantitative electric method, and not the qualitative photographic method of Becquerel. Pierre set up for her, in his engineering College (École municipale de physique et chimie industrielles), the necessary experimental equipment.

Marie established that Thorium also emitted such rays (March 1898). She decided to look not only at pure uranium, but also at uranium minerals; she discovered that pitchblende is more active than uranium and suspected the presence of a yet unknown very active element. At this stage, Pierre decided to join forces with Marie in order to find and separate this element.
In this set-up, a piezoelectric quartz Q, submitted to a certain weight \( \pi \), delivers a certain charge compensating exactly during a certain time (to be measured) the charge delivered by an ionization chamber containing the radioactive material to be studied. The electrometer E allowed this compensation to be monitored.

They developed a new method in chemistry: to perform a succession of chemical reactions and crystallizations and to follow always the most radioactive portion.

In July 1898, they discovered Polonium. In the title of their publication appears for the first time the term “radio-activity”.

In December 1898, they discovered Radium and the physicist E. Demarçay observed a new spectral line corresponding to this new element.

In December 1903, Henri Becquerel, and Pierre and Marie Curie received the Nobel prize for Physics.

Pierre Curie obtained, thanks to the help of members of the science Academies of Paris and Vienna, hundreds of Kg of residues from the treatment of St-Joachimsthal (Bohemia) uranium minerals. Marie started on the very hard chemical work, trying at first to separate Po, and then Ra, which appeared “easier”. In 1901, they decided to work more independently. Marie devoted herself mainly to chemistry: she wanted to
separate pure radium and measure its atomic weight. Pierre devoted himself to physics and to the applications of radioactivity.

In 1901, in Germany, a dentist, Walkhoff, wished to radiograph his teeth with radium radiation, but his gums got burned! Pierre Curie was informed and he tried it out on his own arm, and he also suffers from a severe burn. He then approached Dr. Danlos at the Saint-Louis hospital in Paris and offered to prepare Ra sources in order to treat incurable skin diseases. These were the first medical applications of the radiations of radioactive elements. In 1903, P. Curie and A. Laborde established that radium continuously releases heat. This means that radioactivity is heating our planet Earth. Whereas P. Curie thinks that the energy released in radioactivity is drawn from the environment, in 1902, in Montreal, E. Rutherford and F. Soddy discover that radioactivity is a spontaneous atomic transformation, a conclusion which P. Curie only accepted in 1904.

Pierre Curie died in a street accident in 1906. Marie decided to carry on their common work.

Marie Curie was the first woman professor at the University of Paris. In 1910, together with A. Debierre she isolated pure Radium and measured its atomic weight. She prepared the international standard of radioactivity. In 1911, she received a second Nobel prize, this time for Chemistry.
Radioactive contamination of the laboratory notebooks of Sir William Ramsay and Frederick Soddy: historical and radio-archaeological perspectives*

Neil Todd
University of Manchester
2010

SUMMARY

INTRODUCTION

METHODS

Materials
Apparatus
Procedure

RESULTS

Laboratory Notebooks of William Ramsay
Laboratory Notebooks of Frederick Soddy
Radioactive Letters
Gamma Ray Analyses

HISTORICAL ANALYSES

Isenthal’s Shop and the Production of Helium from Radium, UCL 1903
Later Contamination in the Soddy Notebooks
Later Contamination in the Ramsay Notebooks.

GENERAL DISCUSSION

Historical and Scientific Context in 1903
Technological Development
Technology Transfer

CONCLUSIONS

REFERENCES

* This article has been re-formatted and repaginated from the original - any consequent errors are mine - Editor
SUMMARY

In this paper an account is given of a radiological survey and gamma ray analysis of the laboratory notebooks of Sir William Ramsay, held at the University College London archives, and of Frederick Soddy held in the Bodleian Library at Oxford*. The Ramsay notebooks had previously been surveyed and four such notebooks were identified as being contaminated and held in a box separated from other material. Within the Soddy papers 46 notebooks were surveyed. The notebooks were initially scanned for residual radioactivity with a sensitive Geiger counter and the activities recorded. Selected items from both sets were further analysed by means of a NaI gamma-ray spectrometer for the purpose of radioisotope identification. Both the Ramsay and Soddy notebooks show significant contamination from the summer of 1903 when they were working together at UCL on the production of helium from radium. Both also show later significant (>100 cps) contamination events, Soddy’s in 1905 and Ramsay in 1910. Within the Soddy papers documents which were not used to record experimental laboratory data did not show any contamination, with one exception (some press cuttings from the Times from 1903). The gamma-ray analysis indicated that all of the contamination was due to radium (Ra226). In conjunction with radiological data from buildings and apparatus, these data, as well as data from other surveys, it is argued, provide an insight into some key events in the history of science. Of particular interest is the transfer of technology developed by Ramsay and Soddy for manipulation of radium emanation to the Rutherford school at Manchester and the propensity for these methods to give rise to radioactive contamination.

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INTRODUCTION

The recent discovery of radioactive contamination at Rutherford’s old Manchester laboratory has initiated an interest in the origins and use of the substances that Rutherford possessed at Manchester (Todd 2009a)*, and more generally in the use of radioactive substances in the early part of the 20th century. Although potentially representing a health risk the contamination can also be considered to be an invaluable form of archaeological data which may be used to reconstruct laboratory practise from the early days of nuclear physics. Some progress has been made in reconstructing the Manchester situation during Rutherford’s time and a preliminary account of this is given in Todd (2008). Further surveying of the Rutherford papers held at the University of Cambridge Library has indicated a well-defined historical pattern of contamination which can be correlated with the laboratory contamination (Todd 2009c). In addition to the contamination associated with Rutherford, a question naturally arises whether similar contamination may be found in other laboratories and notebooks from those of his co-workers and contemporaries.

Two such contemporaries or co-workers were the chemists Sir William Ramsay and Frederick Soddy. Although a controversial figure in the Rutherford biographies (e.g. Eve 1939; Campbell 1999), Ramsay’s discovery of the noble gasses and the associated development of the necessary technology to manage tiny quantities of gas had a significant influence on the course of Rutherford’s work. They were also to an extent competitors for the scarce and precious resource of radium in the early years of the 20th century. Frederick Soddy collaborated with Rutherford at Montreal between 1900 until 1903 with a major outcome being the articulation of the theory of successive transformations to account for radioactive decay. A natural issue which arose from this work was the production helium from radioactive decay and it made sense from Soddy’s perspective to work with Ramsay, the discoverer of helium, on this question. In the spring of 1903 he began a series of experiment at Ramsay’s laboratory at University College London which continued until his departure the following year. During the course of this fateful year, cheap radium suddenly became available, thanks to the efforts of the German chemist Friedrich Giesel. The careers of all three protagonists, Rutherford, Ramsay and Soddy, converged during the summer of 1903 after Soddy discovered by accident one day that radium could be purchased from the high street. He happened to be walking down Mortimer Street close to UCL and chanced upon Isenthal’s shop which was advertising radium for sale.
The radium which was purchased that day, and subsequently from the manufacturer played an important part in the Ramsay/Soddy experiments, and in Rutherford’s work, but such quantities of concentrated radium being novel at that time, issues of contamination were not foremost in their proceedings. Subsequent to 1903 Ramsay, Soddy and Rutherford all acquired further quantities of radium. Ramsay in particular obtained on loan from the Austrian government about 300 mg of radium salt. The aim of the survey was to look for evidence of contamination from the Isenthal and later radium and to correlate this with scientific activities in progress at this time.

* Further information concerning Manchester can be obtained at www.rutherfordbuilding.blogspot.com
METHODS

Materials

The William Ramsay papers held in the UCL archives consist of 65 items catalogued under class-mark RAMSAY/1-65. The laboratory notebooks are catalogued as items 22-31. These had been previously surveyed by the UCL Radiological Protection Officer and four notebooks were identified as being contaminated. These were item 26 “Notebook V: The rare gases. The separation of neon from argon and helium; work with Soddy on radon, 1899-1908”, item 28 “Notebook VIA: Miscellaneous notes (much on the work of students), Oct. 1907 – Jun. 1912”, item 30 “Notebook VII: Work with AEC Egerton on transmutation; notes on war work, 1912 – 1914” and item 31 “Notebook VIII: Helium from American natural gas as a fuel for airships; experiments on zinc sulphide for use with radium as luminous paint, Apr. – Dec. 1915”. All four notebooks are kept in a separate box which is clearly marked “Radioactive” with appropriate safety precautions provided. Measurements were limited to these four items.

The Frederick Soddy papers held in the Bodleian Library are classified under the class-mark CSAC 14.8.74. The Laboratory notebooks are held in two sections CSAC14.8.74/IV 46 – 92, contained within four boxes, and CSAC 14.8.74/IV 185 – 215. The first group were bundled by Soddy himself in 1956 and the second group were acquired subsequently from the Inorganic Chemistry Laboratory. The first group contains the notebooks covering his periods at Montreal and UCL. The second group covers period from 1905 at Glasgow but mostly concern work at Oxford. Although both sets potentially contain contaminated items, given the limited time available only the first group were surveyed. The second group will be surveyed on a future visit.

Apparatus

It was expected that any radioactive contamination found would be from the three natural series, i.e. uranium-radium (U238), thorium (Th232) and actinium (U235), but likely be at a low-level. Two sets of apparatus were used for the survey and analysis.
The initial scan was done with a Mini 900 Ratemeter Model EP15, chosen as it is designed to be sensitive to low-level contamination. This instrument should therefore be able to detect the expected low-levels of contamination from the natural series. It has a thin mica window which allows the passage of -particles above 3 MeV energy, and thus should detect -emission from each of the alpha emitters within the natural series (e.g. Ra226, 5.5 MeV; U238, 4.2 MeV; Th232, 4.0 MeV). The EP15 is particularly recommended for soft monitoring, and should therefore detect -emission from radio-lead (Pb210), a decay product of radon from the radium series. The instrument has a maximum sensitivity to photon energy in the range of 30 – 80 keV (peak 50 keV) and therefore would be sensitive to the -emission from Pb210 (46 keV).

For those items selected for further analyses a gamma-ray spectrum was measured with a Berthold LB125 Gamma Analyser. This is designed on a 1.5” by 1.5” NaI Crystal and photo-multiplier sensitive to the range of about 10 – 2200 keV. The ADC allows for 512 channels at 10 bit resolution. The LB125 was interfaced via an RS232 serial port to a laptop PC running the LB125 Spec Software Version 1.3. The software allows for control of data capture and spectrum analysis for peak identification. Calibration was done with a Cs137 source.

Procedure

All measurements were made in the Manuscript Reading Room of the Bodleian or the UCL Archives. A table was set aside and covered with a protective sheet to prevent possible spread of contamination onto the table. For the initial survey all the documents were handled with gloves mainly with the left hand, while the G-M tube from the Mini 600 was handled with the right hand, to minimize contamination of the monitor. As most of the documents were kept in boxes or envelopes each was scanned initially to check for highly penetrative radiation which would indicate that the box or envelope contained contaminated items. If there was any such indication the items were handled more carefully. For each item within an envelope or box the front and back were scanned first and then systematically through each of the pages. If the document showed any sign of activity above background the hot-spots, if any, were identified and recorded. For each such hot-spot a range of activity was recorded at a distance of about 1 cm.
For items with higher contamination some indication of the nature of the radiation was estimated by its powers of penetration. However, positive identification was only possible by means of gamma-ray analysis. Items selected for further analysis were either supported on a book-rest so that the NaI detector could be brought in close horizontal proximity to the hot-spot or if convenient the item was covered with sheets of paper except for the hotspot and the detector supported vertically above the hotspot in close proximity. For the most contaminated items several measurements were made over different energy ranges in order to obtain gamma counts of sufficient magnitude to enable reasonably accurate calculations from a peak analysis, e.g. to compare the peak counts of Ra226 and Pb210. Where only identification was required the count was continued only until a positive identification could be made.

After each session the gloves and table were checked for contamination. If after a session there were signs of residual contamination the glove was disposed of. As a matter of routine hands were washed after each session. The background level was noted before the start of measurements and after measurements in order to assess if instrument contamination had taken place, e.g. from radon.

RESULTS

Laboratory Notebooks of William Ramsay

Of the four notebooks which were previously identified as being contaminated the first two, Vol. V and Vol. VIA/1, were the most substantial in terms of the size, experimental material contained within and significance of contamination. Within each notebook specific episodes of contamination can be identified with definite dates, description of the experimental procedure and radioactive substances used. Details of activities measured are given in Table 1 below.
Table 1 indicates that there were at least two episodes of contamination within Volume V, the first occurring during Soddy’s period at UCL in July 1903 (40 - 60 cps), and the second a year later in July 1904 (800 – 1 k cps). A more significant episode occurs in Volume VIA/1 dating from around February 1911. Volumes VII and VIII are kept together in an envelope and an activity of 20-30 cps can be detected from outside the envelope. Within these two the largest activity, 80 – 100 cps, can be found in Volume VIII dating from October 1915.
Laboratory Notebooks of Frederick Soddy

As noted above, the notebooks which form the group CSAC 14.8.74/IV 46 – 92 were systematically surveyed. These are held in five boxes covering dates from 1894 to 1919; Box 1 (46 – 59), Box 2 (60 – 66), Box 3 (67 – 73), Box 4 (74 – 84) and Box 5 (85 – 92). A large group of these were non-experimental mathematical notebooks, particularly the notebooks of Box 1, which were all at background activity. Of the other notebooks the vast bulk were either at background activity or no greater than 0.5 – 2 cps. There were only two notebooks with activity >2 cps, notebooks 63 and 75, details of which are given in Tables 2a and 2b.

Table 2a: Activity measurements from Soddy’s Notebook 63.

<table>
<thead>
<tr>
<th>Class-mark</th>
<th>Description</th>
<th>Page</th>
<th>Date</th>
<th>Activity (cps)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>1900; 1920; 1955</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Press cuttings from the Times reporting on the Soddy/Ramsay Expts.</td>
<td>22</td>
<td>July 20th 1903</td>
<td>5 – 8 (centre)</td>
<td>'pps. 1 - 23 contain notes made by Soddy 1900 – 1903’</td>
<td></td>
</tr>
<tr>
<td>Press cuttings from the Times reporting on the Soddy/Ramsay Expts.</td>
<td>23</td>
<td>Aug 13th 1903</td>
<td>8 – 10 (centre)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 2b: Activity measurements from Soddy’s Notebook 75.

Location of specific contamination site indicated in bold.

<table>
<thead>
<tr>
<th>Class-mark</th>
<th>Description</th>
<th>Page</th>
<th>Date</th>
<th>Activity (cps)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>CSAC 14.8.74/IV</td>
<td>Notebook 75</td>
<td>9</td>
<td>May 6th 1903</td>
<td>20 – 30 (top left)</td>
<td>Rear of book reads 'March 1903. Chemical Laboratory University College, Gower Street'</td>
</tr>
<tr>
<td></td>
<td></td>
<td>16</td>
<td>June 25th 1903</td>
<td>20 - 40 (bottom right)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1st Expt with Soddy’s 20 mg</td>
<td>18</td>
<td>July 8th 1903</td>
<td></td>
<td>'App put together to test gas occluded in the 20 mg of new RaBr2... The 10 mg from Isenthal is in its original bulb'</td>
</tr>
<tr>
<td></td>
<td></td>
<td>19</td>
<td>July 9th 1903</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2nd Expt with Rutherford’s 30 mg</td>
<td>22</td>
<td>July 11th 1903</td>
<td>50-70 (bottom right)</td>
<td>'A second quite new app was built up to repeat observations with 31.8 mg of RaBr2 kindly lent by Rutherford'.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>27</td>
<td>July 19th 1903</td>
<td>100 – 150 (bottom right)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Eman from 20+30 mg tested for He.</td>
<td>29</td>
<td>July 20th 1903</td>
<td>4 - 5 (left) 150 – 200 (right)</td>
<td>Contamination is Visible on these pages</td>
</tr>
<tr>
<td></td>
<td></td>
<td>30</td>
<td>July 21st 1903</td>
<td>100 – 150 (left) 80 – 100 (right)</td>
<td>Contamination is Visible on these pages</td>
</tr>
<tr>
<td></td>
<td></td>
<td>31</td>
<td>July 22nd 1903</td>
<td>100 – 150 (left) 5 – 10 (right)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>81</td>
<td>June 12th 1905</td>
<td>100 – 150 (bottom right)</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>82</td>
<td>June 20th 1905</td>
<td>200 – 400 (left) 10 – 20 (right)</td>
<td></td>
</tr>
</tbody>
</table>
Notebook 63 indicates a single contamination episode occurred in July/August 1903 (5-20 cps) in the sequence of pages which are noted as by Soddy as 'This END. Notebook began in 1900. Abstracts and References of Radioactivity Papers.' At the location of the contamination Soddy has included some press cuttings from the Times reporting the news of Ramsay and Soddy’s experiments. Notebook 75 show evidence of three distinct contamination episodes. The first dating around May 6th 1903 (20 – 30 cps), the second around July 20th 1903 (150 – 200 cps) and the third around June of 1905.

Radioactive Letters

In a previous survey of the Rutherford papers it was found that a number of letters were also active, the largest number being letters from Soddy. As these are relevant to this survey these results are reproduced here in Table 3.

Table 3. Radioactive letters contained in the Rutherford papers.

<table>
<thead>
<tr>
<th>Author</th>
<th>Classmark</th>
<th>Date</th>
<th>Hot-Spot Activity (cps)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F Soddy</td>
<td>S98, S110, S112 – 115, S118</td>
<td>13th July, 1903 to 16th Mar, 1904</td>
<td>1 - 10</td>
</tr>
<tr>
<td>JJ Thomson</td>
<td>T22, T27</td>
<td>22nd Aug, 1903; 1st Oct, 1905</td>
<td>2 - 10</td>
</tr>
<tr>
<td>F Giesel</td>
<td>G80</td>
<td>14th Mar, 1904</td>
<td>1 - 3</td>
</tr>
<tr>
<td>W Ramsay</td>
<td>R8</td>
<td>22nd Nov, 1907</td>
<td>0.5 - 2</td>
</tr>
<tr>
<td>M Curie</td>
<td>C106</td>
<td>30th May, 1913</td>
<td>1 - 3</td>
</tr>
<tr>
<td>W Lantsberry</td>
<td>L21</td>
<td>6th Apr, 1915</td>
<td>8 – 10</td>
</tr>
</tbody>
</table>
Gamma-Ray Analysis

A number of the most contaminated pages from within the Ramsay and Soddy items were selected for a detailed analysis. These were notebooks V and VI of the Ramsay papers, and notebook 75 from the Soddy collection. The spectra were measured over 2000 keV and 500 keV ranges, illustrated in respectively Figures 1 and 2.

**Figure 1.** Gamma-ray spectrum from contamination found in Ramsay and Soddy notebooks. The four cases were (i) volume V page 318, dated June 20th 1903, (ii) Ramsay volume V, page 396 dated July 21st 1904, (iii) Ramsay volume VI, page 128, dated Feb 18th 1911 and (iv) Soddy notebook 75, page 29, dated July 20th 1903. The energy range was 2000 keV and the count was taken over 1000 s. Peaks for isotopes from Ra226 and daughters Pb214, Bi214 and Pb210 are clearly identifiable.
It is clear from the examination of the gamma-ray spectra that all of the radioactive contaminants analysed are radium (Ra226) and daughters. It is of interest to consider what further information may be obtained from the shape of the spectra. Apart from a baseline shift due to differences in overall activity, there are observable differences in the relative heights of some of the individual peaks. For example for the 2000 keV spectra the peak ratio Pb210/ Ra226 varies between 0.92 to 1.55 and Pb214/ Ra226 between 0.57 and 1.1. For 500 keV range these are 1.47 – 1.56 and 0.96 – 1.14 respectively. The sources of this variability are likely a combination of interference from background activity, differences in age and in rate of diffusion of radon away from the hotspot. At the lowest activity the Pb210 and Ra226 peaks although present are less well-defined, suggestive of background interference. There appears to be a trend for the Pb214/ Ra226 ratio to increase with activity, again suggestive of interference at the lowest
activity.

In a closed system the ratio Pb210/Ra226 will be precisely related to the age of the radium since fractionation. Given the large difference in half-life of the two isotopes, 22.7 and 1600 respectively, we may assume that the amount of Ra226 is constant so that the age of radium is given by

$$t = -\frac{1}{\lambda} \ln \left( 1 - \frac{A_{Pb210}}{A_{Ra226}} \right),$$

where APb210 and ARa226 are the measured activity levels of Pb210 and Ra226 respectively and is the decay constant for Pb210. However, for an open system a proportion of the Pb210 will be lost due to radon diffusion. If the ratio of lost and retained Pb210 is an unknown value, then the relation for age will be modified to

$$t = -\frac{1}{\lambda} \ln \left( 1 - \frac{A_{Pb210}}{A_{Ra226}} (1 + \alpha) \right).$$

We may thus approach the radium samples from either assuming $\alpha = 0$ and estimating age, or by taking the age given by the dates in the notebooks and estimate the proportion of lost Pb210. In order to investigate these properties of the Ramsay and Soddy radium, estimates were made of APb210 and ARa226 for each of the three cases shown in Figure 2. Peak areas (PA) were measured using the method of Gilmore (2008) by integrating over the each peak to obtain the total area (TA) and subtracting the background area (BA). The activity was then taken to be given by

$$A = \frac{PA}{p \cdot T \cdot \varepsilon(\nu)} = \frac{TA - BA}{p \cdot T \cdot \varepsilon(\nu)}$$

where $p$ is the probability of a gamma emission, $T$ is the count time and is the counter efficiency as a function of photon energy (assumed to be 1 here). The estimates are given in Table 4, along with an estimate from contamination found in papers of Norman Feather for comparison (Todd 2010). The figure in parentheses is due to uncertainty in the nominal age of the radium, it may have been Rutherford’s old Vienna radium from 1908.
Table 4. Estimated activity ratios, radiometric dates or loss rates from the Ramsay/Soddy papers.

<table>
<thead>
<tr>
<th>Case</th>
<th>APb210 (Bq)</th>
<th>ARa226 (Bq)</th>
<th>Ratio</th>
<th>Radiometric Age (yrs)</th>
<th>Nominal Age (yrs)</th>
<th>Loss Rate = 100%/(1+ )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soddy 75, 29</td>
<td>77 - 115</td>
<td>50 - 118</td>
<td>0.91 – 0.98</td>
<td>79 - 128</td>
<td>106</td>
<td>-2 – 6%</td>
</tr>
<tr>
<td>Soddy 75, 82</td>
<td>87 - 107</td>
<td>85 - 117</td>
<td>0.74 – 0.91</td>
<td>44 - 79</td>
<td>104</td>
<td>5 – 23%</td>
</tr>
<tr>
<td>Ramsay VI, 128</td>
<td>1102</td>
<td>1633</td>
<td>0.675</td>
<td>37</td>
<td>99</td>
<td>29%</td>
</tr>
<tr>
<td>Feather 13/5</td>
<td>1126</td>
<td>1577</td>
<td>0.71</td>
<td>41</td>
<td>80 (101)</td>
<td>22% (?)</td>
</tr>
</tbody>
</table>

The above method is probably reasonably accurate for high activity sources, but less so for low activity sources where noise in the spectrum curve makes estimating BA prone to error. Uncertainty in these cases is indicated in Table 4 by a range of values. The error for the Soddy cases is likely quite high, so that for example some of the ratios are > 1, which is not physically possible, and some age estimates are greater than the nominal age, suggestive that the Pb210 count is too high (possibly due to there being additional Pb210 present). Nevertheless, the outcomes of the analysis are suggestive that if greater accuracy could be achieved, that some useful procedures could be developed for approximate dating of archival and archaeological contamination. For example, from a statistically larger sample of archival cases, where the dates are known, it would be possible to obtain a range of values for loss rate which could then be used to correct the radiometric age where dates are not known.

HISTORICAL ANALYSES

The above survey of archival contamination in the notebooks and papers of Ramsay and Soddy indicates a small number of contamination episodes between 1903 and 1911. The most extensive episode is the earliest one which occurs during the summer of 1903. Remarkably, evidence of this episode can be found on four independent sets of documents including Ramsay’s volume V, Soddy’s notebooks 63 and 75 and Soddy’s letters to Rutherford from summer 1903. There are a small number of isolated later cases, but the most significant is one which occurs in the Ramsay’s notebook VI between 1910-11. In what follows I place the 1903 episode and the later Ramsay episode in their historical context.
There can be little doubt that the 1903 episode was related to the sudden availability of concentrated quantities of radium care of the German chemist Friedrich Geisel (Fricke 2001). Prior to this, only weak preparations of radium chloride were available through the Curies. In the spring of 1903 Soddy had moved from Montreal, where had been working with Rutherford, to London to work with William Ramsay at UCL on the production of helium from radium. Sometime after starting at UCL in March 1903 Soddy had chanced upon Isenthal’s shop on Mortimer Street, about 20 minutes walk from UCL, which was advertising Giesel’s radium.

“Our trouble here was the same as in Montreal. We had a quite insufficient amount of radium for our investigations. Then, by the most extraordinary chance, the whole future prospect was changed.... I was walking along Mortimer St off Upper Regent
Street, in London one day when, casually looking through Isenthal's window, I saw advertised something I could not credit to be true: 'pure radium compounds on sale here' ... I learnt from the salesman that Professor Giesel in Germany,..., had started manufacture of radium compounds on a commercial scale in the Chinin Fabrik at Brunswick. He used residues left after the extraction of uranium from the Pitchblende found in the old state silver mine of St Joachimsthal in Bohemia. Isenthal had quickly taken advantage of this supply.

At that time one could buy radium only from the French factory by favour of the Curies. Here it was to be bought in a London shop at some eight shillings a milligram of pure radium bromide. ...

I hurried back to UCL to tell Ramsay of my treasure. Looking at it in the dark with an X-ray screen would have convinced the investigator that it was genuine. Ramsay, to my absolute horror, stuck in a moistened platinum wire, removing a large fraction of my few milligrams and held it in a Bunsen burner to see if it gave the red carmine flame characteristic of radium free from barium. It was the only time I have seen that flame and I never want to see it again! It made me almost ill to see this treasure treated so profligately, besides permanently spoiling the laboratory for any delicate radioactive work. Ramsay at that time had little experience of radioactivity.” [Soddy 1957, quoted in Howarth (1958)]

This passage clearly indicates that a contamination event occurred during 1903 at Ramsay’s laboratory, shown in Figure 3. It is of interest to attempt to correlate the 1903 archival contamination with the time line of events that transpired during this period, and this is provided in Table 5 below. Although we do not have an exact date for Soddy’s visit to Isenthal’s shop, it is apparent that the amount, which is referred to on July 9th in NB 75, was 10 mg. On this date a further amount of 20 mg is noted, which had been delivered from the manufacturer Friedrich Giesel of Brunswick*. It was this further quantity of radium that was used successfully for the helium production experiments, rather than the original Isenthal 10 mg. We may infer that Soddy’s discovery of Isenthal’s radium probably occurred sometime in May and before June 10th, when Ramsay notes that Soddy has his own radium. The principal 1903 archival contamination occurs well after this between 19th – 22nd July. This indeed occurs after a first experiment to test for helium production with the further 20 mg on July 9th and a second experiment on July 11th making use of a third quantity of radium, about 30 mg, which had been purchased by Rutherford from Isenthal’s on a visit to London that July.

“I told him immediately on his arrival at UCL about my find at Isenthal’s and together we lost no time in walking round to Mortimer St. I must say he experienced the same astonishment and joy that I had experienced when confronted with radium
bromide for eight shillings a milligram some weeks before. He was absolutely bowled over and became as excited as a school boy over the coming holidays. With thirty precious milligrams of pure radium bromide we bounded back to Gower Street and we both immediately repaired to the dark room with some metal foils and a bit of X-ray screen. The effect was terrific; it was like a person born blind suddenly being given sight, for though R had made a special study of the Becquerel rays, this was the first time he had ever seen them. All his work had been done by the ionization method with substances far too feebly radioactive to light up the X-ray screen. Now he had a visual demonstration of what he had found out in the dark - so to speak.” [quoted in Howarth (1958)]

*On this point Maurice Travers appears to be in error in his belief that the 20 mg of Geisel radium delivered in early July was the same radium which Soddy refers to in his anecdote. Travers though had not had an opportunity to review Soddy’s own notebooks.

According to Campbell (1999) the visit by Rutherford actually took place during the first experiment on July 9th. That this experiment was being conducted by Soddy apparently came as somewhat of a surprise to Rutherford and was a source of some resentment. He nevertheless subsequently loaned his 30 mg for the second experiment. The first sign of contamination from this time is a letter from Frederick Soddy dated 13th July 1903 (S98) which has a hot-spot on the top of 8 – 10 cps (identified as Ra226). This was two days after the start of the second experiment and in it he thanks Rutherford for the loan.

“If you received my wire you will have heard the results of the second experiment with your radium. … It is a veritable triumph and we are very grateful to you for making it possible. Your radium weighed 31.8 mg and I hope soon to hear from the manufacturers when I shall get my 30 mg from them.”

The bulk of the 1903 contamination then, takes place after completion of the two helium from radium experiments and during follow up experiments from 17th to 22nd July when Ramsay and Soddy attempt to measure the helium production from emanation and the spectrum of radium emanation itself making use of both the 20 and 30 mg radium samples. The extraction and merging of emanation from two sources in solution was undoubtedly a more complex procedure, and this may have contributed to the contamination. The Summer vacation was fast looming and this may have injected some haste into the proceedings.
Table 5: Correlation of activity measurements from the Ramsay and Soddy notebooks.

<table>
<thead>
<tr>
<th>Date</th>
<th>Event</th>
<th>Soddy NB 63 &amp; 75</th>
<th>Soddy Letters</th>
<th>Ramsay V</th>
</tr>
</thead>
<tbody>
<tr>
<td>March</td>
<td>Soddy arrives at UCL</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>March 7th</td>
<td>Soddy writes to Rutherford. “I had an interview with Ramsay giving a short account of our results”.</td>
<td></td>
<td>S 96, Eastbourne</td>
<td></td>
</tr>
<tr>
<td>May 6th</td>
<td>“Prof Trator’s Ra in bulb”</td>
<td>20-30cps, NB75, p9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>May 9th</td>
<td>Ramsay’s apparatus to show Ra in bulb heated in bulb to give off gases for purification (see Figure 2)</td>
<td></td>
<td></td>
<td>p310</td>
</tr>
<tr>
<td>May-June</td>
<td>Soddy finds Isenthal’s shop during this time, buys 10 mg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>June 10</td>
<td>Ramsay notes that Soddy has his own RaBr2</td>
<td></td>
<td></td>
<td>p311</td>
</tr>
<tr>
<td>July 9th</td>
<td>Soddy notes “The 20 mg RaBr2 from a subsequent letter of Muller-Uri had been prepared in solid state some 2 ½ months. ... Apparatus put together to test the gas ... in the 20 mg of new RaBr2 from Muller-Uri ...The 10 mg from Isenthal in its original bulb.”</td>
<td>NB 75, p18</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Rutherford visits Soddy at UCL</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>July 11th</td>
<td>“A second quite new app was built up to repeat observations with 31.8 mg of RaBr2 kindly lent by Rutherford”.</td>
<td>NB 75, p22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>July 13th</td>
<td>Soddy informs Rutherford of the results of the second experiment. “It is a veritable triumph”</td>
<td></td>
<td>S98, Notting Hill 5 – 8 cps</td>
<td></td>
</tr>
<tr>
<td>July 19th</td>
<td>Spectrum of Eman using 20 + 30 mg of Geisel radium. Soddy’s hand</td>
<td>40 – 50 cps p317</td>
<td></td>
<td></td>
</tr>
<tr>
<td>July 20th</td>
<td>Press cuttings from the Times reporting on the Soddy/Ramsay Expts.</td>
<td>5 – 8 cps, NB 63, p22</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Eman from 20+30 mg tested for He</td>
<td>150 – 200 cps NB 75, p29</td>
<td></td>
<td>50 – 60 cps p318</td>
</tr>
<tr>
<td></td>
<td>Spectrum of Eman using 20 + 30 mg of Geisel radium. Ramsay’s hand</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>July 21st</td>
<td></td>
<td>100 – 150 cps</td>
<td>20 – 30 cps</td>
<td></td>
</tr>
<tr>
<td>July 22nd</td>
<td></td>
<td>NB 75, p30</td>
<td></td>
<td>p321</td>
</tr>
<tr>
<td>July 28th</td>
<td>Ramsay &amp; Soddy (1903) ms is received by the RS</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>Vacation begins and radium put in bulbs attached to pump for storage</td>
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<td></td>
</tr>
</tbody>
</table>
Returning to the flame test contamination incident, there appears not to be a record of this in the Ramsay and Soddy notebooks. There is a 20-30 cps hot spot which occurs in a page dated May 6th from NB75, but this seems too early for the Isenthal radium. However, if the flame test was carried out spontaneously by Ramsay it is unlikely that the laboratory notebooks would be open or close by the apparatus so that they would not show evidence of it. It seems curious though that Ramsay and Soddy do not refer to the use of the original Isenthal 10 mg, although it is possible it was used in the earlier unsuccessful experiments. Prior to the arrival of the 20 mg, Ramsay and Soddy in their later paper of April 1904 only talk of use of an impure radium chloride. If, as Soddy suggests in his anecdote, a “large fraction of my few milligrams” was wasted, the original 10 mg may have been reduced to as little as 5 mg and it may have been considered too small a quantity to be of practical use.

After the end of the Summer term, and over the following months, during which time Rutherford and Soddy had met again at the British Association meeting at Southport in September, there was a sequence of letters from Soddy to Rutherford which also show hot-spots of activity. Most of these concern the purchase and postage of further consignments of Giesel’s radium and apparatus for its use. On October 12th Soddy was able to tell Rutherford that:

“We are in luck for Giesel sent us his pure RaBr2 this morning & fine stuff it is too. He only charged us 12 Marks per mg which is less than you paid Isenthal. Tomorrow when I go to the lab I will weigh out 31.8 mg and send this to you with this letter…”

Thus by the end of October Rutherford had had his loan of 30 mg returned. Although he may have had some lingering resentment concerning the Ramsay and Soddy experiments, as we discuss later, he did benefit from the technological advances made during them.

Later Contamination in the Soddy Notebooks

Other than the hotspots found in the Soddy/Rutherford letters from this period, there was also a hotspot found in NB63 associated with a second press cutting dated August 13th. This was on the page opposite the July 20th press cutting and may have been contaminated on the same occasion.
The only other contamination to be found was from NB75 on pages dated June 1905. The gamma-ray analysis indicated that this radium was similar to the 1903 radium so it is possible that it did actually occur in 1903. It is also possible that there was interference in the measurements, i.e. rays from one hotspot may have confounded the measurements of the other. Further more careful measurements will be required to resolve this.

*Later Contamination in the Ramsay Notebooks.*

In contrast to the Soddy notebooks, the Ramsay notebooks contain two other more significant contamination episodes, from 1904 in Volume V and from 1910/11 from Volume VIA. Both of these are visible on the pages where they occur.

**H and O$_2$ produced by Emanation, July 1904**

About one year after the contamination in July of 1903, the next significant event occurs in Ramsay’s volume V on pages 393-397 with the peak contamination on page 396 dated July 21st where counts of up to 1000 cps can be recorded. By this time Soddy had obtained a lectureship in Glasgow and was no longer working at UCL. It is apparent from reading the notes on this page that the contamination, which is visible, occurred during an experiment to measure the production of hydrogen and oxygen dissociated from water by the actions of radium and radium emanation.

**Fractionation of RaBr$_2$ using 328.5 mg RaBaBr$_2$**

By far the most significant contamination in the whole set of notebooks occurs at the end of 1910 early 1911 in volume VIA. The highest readings, which exceed 2000 cps, can be made on pages 126 – 129, dated Feb 14th – 18th 1911. The notes on these pages record that Ramsay was carrying out a fractional recrystalisation of a large sample of 328.5 mg RaBaBr$_2$. By 1911 Ramsay had acquired much larger quantities of radium, not least a large sample from the Austrian Government in 1908 which was intended for sharing with Rutherford. The fractionation procedure was inherently more prone to accident and it is clear from Ramsay’s notes that a spill and attempted recovery by pipette did indeed occur on these pages.
GENERAL DISCUSSION

The survey of the Ramsay and Soddy documents has indicated a small number of well-defined contamination episodes which can be correlated with historical information relating to specific scientific events. The most significant episode historically, although not radiologically, was the contamination dating from the Summer of 1903. A number of more general issues arise from this episode which I discuss below.

Historical and Scientific Context in 1903

The first issue to consider is the historical and scientific context in which the 1903 Isenthal episode took place. It did not happen in isolation, but would appear to be a sample from a more widespread surge of radioactive contamination events which occurred at this time. We have already noted above that 1903 was a significant year in that it was marked by the sudden availability of cheap radium of high purity, largely due to the work of Friedrich Giesel and the chemical manufacturers Buchler & Co of Brunswick. Through their agent (and glass blower) Müller-Uri, Isenthal’s and other retail outlets were supplied with radium so that it became easily obtainable by general members of the public as well as scientists.

It was this availability that undoubtedly fueled a great demand for the stuff by both amateur and professional alike. As I have documented elsewhere (Todd 2009b), there was in the scientific literature, not least in the journals Nature and Proceedings of the Royal Society, from 1903 an unprecedented flood of articles on radium studies. Whereas Volume 71 of the Proceedings of the Royal Society had published only a single paper recording work with radium, Volume 72 published no less than eight papers in the space of five months from between May and October 1903, and there is a remarkable sequence of four consecutive radium articles between 17th July and 5th August (including a paper by Ramsay and Soddy (1903)). During 1903 a total of nine papers appeared in the Proceedings with the word radium in the title, a number never to be exceeded in any future year.

Rutherford, of course, was a beneficiary of this source of radium and a survey of his papers show that some of the highest levels of contamination, which is present from 1900 until the late 1920s, can be found from 1904, shortly after he received the Giesel radium (Todd 2009c). Although at this time I have not had an opportunity to survey other archives we it would be a good bet to suppose that the contamination found in the Rutherford, Soddy and Ramsay papers is likely present in any surviving documents from other
scientists of this period. Among these contemporaries are the other contributors to Volume 72 of the Proceedings, including William Crookes (1832 – 1919), James Dewar (1842 – 1922), R J Strutt (4th Baron Rayleigh, 1875 - 1947) and W B Hardy (1864 – 1933). Certainly there is plenty of anecdotal evidence to support this view. For example, contamination at the Royal Institution during Dewar’s occupation from a spill of Curie radium is referred to in letters and WB Hardy is known to have received a severe radium burn through careless handling. Further surveying work is required though to substantiate these suspicions.

We may also mention Arthur Schuster, who was also caught up in the great 1903 radium boom like many others at the time. Soddy visited Schuster in Manchester on his way to the British Association Meeting in Southport that year. Undoubtedly, radium was a hot topic of conversation, as it was at the BA meeting. Shortly after Soddy’s visit, Schuster obtained in October of 1903 radium bromide from another London retailer, Armbrechts and Nelson, initially purchasing 20 mg and then another 40 - 50 mg so that he able to write in 1906 that he possessed about 60 – 70 mg. We can be in little doubt that Manchester had experienced its own early episode of contamination prior to Rutherford, and this is supported by evidence that Rutherford had problems keeping his electrosopes charged right from the start of his tenure in 1907.

Technological Development

A second important issue is the technological development for management of radium and radium emanation (radon) which took place in the Summer of 1903 at Ramsay’s laboratory. These developments had a considerable influence on the wider community of radioactivity researchers at this time, not least through the direct transfer of technology to Rutherford in Montreal via Soddy. As well as being of intrinsic interest for the history of science, the technology has implications for the archaeological signature which it might deposit, both in the form of radioactivity and in the form of mercury, large quantities of which were required for the reservoirs.
Figure 4. Sketch of early apparatus for extracting gas from radium, dated May 9th 1903 from page 310 of Ramsay’s Volume V. Radium introduced in solution in a bulb at one end (right) and drawn through by means of Toepler mercury pump at the other end (left). The U-tubes were immersed in Dewars containing either dry ice of liquid air. This appears to be a precursor to the apparatus illustrated in Figure 1 of Ramsay W and Soddy F Proceedings of the Royal Society Vol. 73 (1904), pp. 346-358.

A description of the technological development is given in by Travers (1956). The essential problem which was faced by Ramsay and Soddy was is how to manipulate the minuscule amounts of gas involved and measure their physical properties. This problem was compounded by the fact that radon was continually disappearing by radioactive disintegration. Travers noted that a sketch of apparatus used in early experiments appears in Volume V. This is reproduced above in Figure 4. Essentially the same apparatus is illustrated in Figure 1 of Ramsay and Soddy’s 1904 paper, again reproduced below in Figure 5 here. Gas could either be directly evolved from a solution of radium salt in bulb A, or re-introduced via the inverted siphon, after drawing off and purifying in a separate apparatus. The spectrum of the gas could be measured in the U-tube B.
Figure 5. Early apparatus used by Ramsay and Soddy to measure the spectrum of helium from radium, a development of the apparatus sketched in Volume V. A solution of radium salt was introduced in bulb A and the gas drawn off. Alternatively gas could be introduced at the inverted siphon in a mercury trough from a separate apparatus. The spectrum was measured at U-tube B. Adapted from Figure 1 in Ramsay W and Soddy F Proceedings of the Royal Society Vol. 73 (1904), pp. 346-358.

According to their papers these early experiments were carried out using an impure radium chloride salt and were not successful in obtaining the helium spectrum. As late as July 4th Ramsay had written in a letter that the work was difficult, even though Soddy had a larger amount. It is not clear if this larger amount is the original Isenthal 10 mg or the later 20 mg which was delivered from the manufacturer and noted on July 9th by Soddy. Nevertheless, a number of improvements were made which included use of capillary tubing throughout and use of a micro-spectrum tube, replacing the U-tube of the early apparatus. An illustration of the new apparatus is shown below in Figure 6. In fact at least two versions of this were made, one for Soddy’s 20 mg RaBr2 and a separate one for Rutherford’s 30 mg. The gas evolved from the dissolved salt could either be taken off from a bulb
directly attached to a Toepler pump, as in Figure 5, and introduced via the inverted siphon, or the bulb directly attached to the tube between the siphon A and B. The second alternative, which was employed for Rutherford’s radium, was thought to be less prone to contamination, an issue we return to later.

Figure 6. Later apparatus used by Ramsay and Soddy to measure the spectrum of helium from radium. The gases could either be introduced at inverted siphon A in a mercury trough from a separate apparatus (Toepler pump & radium bulb), as was done with Soddy’s 20 mg, or introduced directly from bulb H, as was done with Rutherford’s 30 mg. Components B + C were used to recombine dissociated hydrogen and oxygen and the remove the water. After the radium emanation was frozen out in the U-tube E helium could be compressed into the spectrum tube F. Adapted from Figure 2 in Ramsay W and Soddy F Proceedings of the Royal Society Vol. 73 (1904), pp. 346-358.

In addition to the above, the Ramsay and Soddy apparatus made use of two other components which were essential to success. The gas which was evolved from radium solution contained mostly dissociated hydrogen and oxygen so it was necessary first to recombine them either by sparking from a Leyden jar and induction coil or by heating by copper wire. Once
recombined as water it was then necessary to remove this by use of phosphorus pentoxide (P2O5), a powerful desiccant. These two components are indicated in Figure 6 as B and C. There tended though to be an excess of hydrogen so additional oxygen was introduced through the inverted siphon. Once these procedures had been carried out the remaining gas was shunted in the U-tube E and compressed by raising the mercury reservoir. As the boiling point of radon is about 211 K, compared with 4 K for helium, and air is liquid between about 60 – 80 K, then any helium present could be separated and concentrated in the micro-spectrum tube F by emersion of the U-tube in liquid air. From the spark spectrum any helium present could be identified by spectroscopy, the yellow D3 line (588 nm) being the most prominent.

In addition to the need to recombine the dissociated H2 and O2 and remove the resultant water, there was also a problem that the radon attacked the tap grease with the production of CO2. The boiling point of CO2 is 195 K, so it would also be frozen out with the emanation (radon). For the helium detection experiments this was sufficient, but if the properties of the emanation were to be investigated it was necessary to remove the CO2.

This was accomplished by reducing the number of taps, and substituting them with mercury traps, and by the use of caustic potash. An example of apparatus which made use of these new developments is given below in Figure 7.
Figure 7. Later apparatus used by Ramsay and Soddy to measure the spectrum of helium and radon, without CO2. The radon-electrolytic gasses were extracted and sparked in a separate apparatus (Toepler pump, radium bulb + spark chamber) and the residue introduced at the inverted siphon. Components B, C and D contained respectively potash solution, solid potash and P2O 5. Adapted from Figure 4 in Ramsay W and Soddy F Proceedings of the Royal Society Vol. 73 (1904), pp. 346-358.

These techniques were further developed by Ramsay and colleagues after Soddy’s departure in the Spring of 1904. However, the critical developments for manipulation of radon had been made by this time. One important feature of the development of apparatus from the May 9th 1903 sketch to the later apparatus described in their 1904 paper, is that the production and manipulation of radium emanation now required two separate pieces of apparatus. As is noted by Travers, one consequence of this was the increased risk of contamination, and it is likely that this was a contributing factor to the 1903 contamination episode.
Technology Transfer

A further consequence of the division of apparatus was that two separate Toepler mercury pumps were now required. In the first piece of apparatus a bulb containing radium in solution was attached to a pump for extraction of radon and electrolytic gasses. In the second, evacuated by the second pump, the mixed gasses were introduced via inverted siphon and after purification the helium or radon were compressed into a capillary tube, spectrum tube or other small receptacle. We know from Travers that Ramsay did possess two pumps, as illustrated in Figure 3. When Rutherford made his visit on 9th July 1903 he would have witnessed in Ramsay’s laboratory these two pieces in action. It is clear from what followed that, in spite of any resentment he may have felt, he was greatly interested in the technological developments. In addition to his purchase of 30 mg of radium bromide we know from Soddy’s letters that he also ordered two Toepler pumps which were duly delivered that year, and these can be seen in the famous photograph of Rutherford taken in his basement laboratory in Montreal in 1905. When he arrived in Manchester in 1907 we know that he did not bring the radium, but we do not know if he also left the pumps behind. Whatever the case, at Manchester he had the benefit of glass-blower Otto Baumbach who would no doubt have been able to build a new set of pumps for him. Figure 8 below shows an example of apparatus used by Ramsay and Collie compared with apparatus used by Rutherford to keep radium in solution.
Figure 8. Examples of Toepler pumps. (a) Apparatus used Ramsay and Collie (1906), from Figure IX.1 in Travers (1956); (b) Apparatus for keeping radium in acid solution and for drawing emanation, adapted from Fig. 107 of Rutherford (1912) Radioactive Substances and Their Radiations. The pump operated by raising or lowering the mercury reservoir. With the stopcock S open, lowering the reservoir would draw gas into the body of the pump T. With the stopcock closed raising the reservoir would shunt gas into the burette B.

On first arrival to Manchester Rutherford was faced with a shortage of radium (Eve 1939); he did not have his Giesel radium and Schuster’s radium was largely unavailable, either sold, in use or perhaps lost. A joint loan to both Rutherford and Ramsay by Academy of Science of Vienna did not result in a satisfactory arrangement for Rutherford, as Ramsay wished to keep it UCL (although for a short while Ramsay sent consignments of emanation by train up to Manchester). After Rutherford obtained his own loan of radium salt from Vienna in February 1908 he employed the apparatus shown in Figure 8 (b) to keep it in solution. Thereafter, the radium emanation could be periodically milked off over mercury for the various experiments he and co-workers were engaged in at Manchester. The availability of a large radium source enabled him to embark on a series of experiments investigating the properties of radium emanation, as well to produce radioactive sources. It is quite clear that the apparatus used for these is directly derived from the Ramsay and Soddy developments, and this is acknowledged by Rutherford.
Figure 9. Apparatus for purification of emanation and transfer to “emanation tube” F, adapted from Fig 1 of “Experiments with the Radium Emanation”, Phil Mag XVI (1908), pp 300-12.

As a first example of this, Figure 9 above shows the apparatus used by Rutherford in his paper “Experiments with Radium Emanation” (Rutherford 1908). After extraction of emanation from solution using the apparatus shown in Figure 8(b), the evolved gas was “sparked down”, to remove O2 and H2, in a second piece of apparatus and then transferred into the apparatus shown in Figure 9 for final purification and measurement. All of the components of this are essentially the same as Figures 6 and 7, except that the sparking is done separately. Burette D is lined to caustic potash to
remove CO2, the U-tube is immersed in liquid air to condense the emanation and uncondensed gasses H2 and He pumped off. Finally, the emanation is allowed to volatilize into tube E and compressed into capillary F, where its volume can be measured. Alternatively the spectrum could be measured if the capillary is exchanged for a spectrum tube, which may be removed to another location for spectroscopic analysis and photography (Rutherford and Royds 1908). As noted above, Rutherford gives clear acknowledgement to Ramsay for this.

“In this work, I have found the methods developed by Ramsay and others for manipulating small quantities of gases of great assistance. See Travers, ‘Study of Gases’”. [Rutherford 1908, p. 74 of Chadwick 1963]

A second example is the apparatus used by Rutherford and Royds (1908) to investigate the effect of emanation on water and the various gases involved, illustrated below in Figure 10. They were particularly interested to determine if neon is produced, as claimed by Ramsay. Again, most of the components are derivative of the Ramsay methods, as is acknowledged in the text. The form is similar to that of Figure 9, but with some additional features. Most notable the use of an exhausted charcoal tube H, which would absorb all gases in air except neon and helium.
As a third example of technology transfer, illustrated in Figure 11, is a complete set of apparatus for keeping of radium and purification of evolved gases for the production of radioactive sources described by Makower and Geiger in their 1912 text book. This text “Practical Measurements in Radioactivity” was written to accompany their training class in radioactivity which all new members of Rutherford’s group at Manchester had to go through. By this means the methods developed by Ramsay and Soddy in 1903, for keeping radium and for extracting and purifying radium emanation, became standardised within the radioactivity community. At the same time as promulgating the standardised procedures, the propensity for these procedures to give rise to associated contamination of the kind found in the notebooks of Ramsay and Soddy from 1903 was also transmitted to the new practitioners. As noted above, every additional manipulation and
division between separate pieces of apparatus could only increase the risk of contamination. Some of the highest levels of historic contamination found at Manchester, along with high mercury contamination, was found in the room used by Rutherford to store radium, and where most of the above described glass apparatus was used (Todd 2008).

**Figure 11.** Apparatus described by Makower and Geiger (1912) for keeping radium and for purification of evolved gases, adapted from Figures 58, 59 and 60 of “Practical Measurements in Radio-activity”. (a) Radium solution bulb attached to Toepler pump, (b) sparking apparatus for removing electrolytic gas and (c) condensation apparatus for final purification and compression into a “radon tube”.
Figure 12. Apparatus use by Rutherford and Royds (1909), adapted from the figure of “The Nature of the particle from Radioactive Substances”, Phil Mag XVI, pp 281-6.

The most famous experiment carried out in the “Radium Room” at Manchester (on the top floor of the old Physical Laboratory (Schuster and Hutton, 1906)) was the Rutherford and Royds (1909) experiment to determine the helium nature of the alpha particle. Once again, we can see that most of the components, illustrated in Figure 12, e.g. the pump, the mercury reservoirs, etc, are derivative from the Ramsay methods. However, there is one development here which is uniquely Rutherfordian and Mancunian. The trick was to use a fine glass tube A, produced by glassblower Baumbach, thin enough to allow particles to pass through, but thick enough to withstand atmospheric pressure. When the thin tube was filled with purified radium emanation, gradually over a few days helium gas in a surrounding outer tube T became detectable spectroscopically after compression into a spectrum tube V.
CONCLUSIONS

An analysis of radioactive contamination found in the laboratory notebooks of William Ramsay and Frederick Soddy confirmed that this was due to radium. Evidence of the earliest episode from the Summer of 1903 is found separately in four documents. This occurred within weeks of pure radium preparations first becoming commercially available in London. At least part of the explanation for the contamination may be due to the division of apparatus between storage and purification and necessary transfer of substance between divisions. The easy availability of radium and methods developed for handling it and associated gases by Ramsay and Soddy had a large influence on Rutherford and his science. At Manchester these methods were employed on a large scale for the production of radioactive sources and passed onto to Rutherford’s many students. Undoubtedly, much of the Manchester contamination from 1908 resulted in the same manner as that which gave rise to the contamination at UCL in 1903.

REFERENCES


The Early Influence of Manchester on the Work of George de Hevesy

Siegfried Niese

Wilsdruff, Germany, former VKTA - Radiation Protection, Analytics & Disposal Inc.

Summary

The Nobel laureate Georg de Hevesy (1885-1966) had studied physics and chemistry and in 1908 he gained his PhD in Freiburg im Breisgau. After two years as assistant in Zurich and Karlsruhe he spent two years, from 1910 – 1912, in Manchester to learn something about radioactivity with Ernest Rutherford. In the very creative atmosphere he learned new techniques and ideas, became interested in research work, and found friends like Moseley, Russell, and Bohr. In this paper I suggest that the stay in Manchester was fundamental for his long and successful scientific career making important discoveries in physics, chemistry, geology, physiology, and medicine.

Introduction

Hevesy’s stay in the Physical Laboratory of the University Manchester headed by Ernest Rutherford who was awarded the Nobel Prize for chemistry in 1908 for the discovery of transformations of elements by the radioactive decay, i was extremely important for his further scientific career. In 1911 Rutherford had shown that the scattering of α - particles by atoms, from work carried out by Hans Geiger and Ernest Marsden, ii could only be explained by a positively charged, extremely small nucleus, with the electrons rotating like planets around the sun at a distance which determined the dimension of the atoms. iii By the time Hevesy arrived at the laboratory, many natural radioactive elements and their radiation and lifetimes were known. He arrived in Manchester to learn something about radioactivity, and was fascinated by the high level of scientific discussions in the laboratory. It will be shown that Hevesy’s stay in Manchester was very important for his further scientific work. The life and work of Hevesy are described in his autobiographical remarks iv, in the Biographical Memoires of Fellows of the Royal Society v and in biographies of Hilde Levi vi, Gabor Pallo vii and Siegfried Niese viii.
Education

George de Hevesy was born on August 1, 1885 in Budapest, the fifth child of a wealthy aristocratic Hungarian Jewish family. After education by private teachers he went to a catholic school of the piarists in Budapest, where some of the teachers had been professors at universities. He studied in Budapest, Berlin, and Freiburg, and in 1908 he was awarded a Ph.D. at the University of Freiburg. In his thesis Hevesy determined the solubility of sodium in molten sodium hydroxide which he found responsible for the reduced yield in the electrolysis of sodium. Because of the growing importance of electrochemical processes in metallurgy he had planned to work in a metallurgical enterprise in Hungary and was not at that point much interested in radioactivity. The head of the Institute of Physical Chemistry in Freiburg, Georg Meyer mainly concerned with electrochemistry but became interested in radioactivity after acquiring some radium. Until 1910 Hevesy was an assistant in the Institute of Physical Chemistry in Zurich, but wishing to understand more about the electron he considered applying to Thompson but he was advised to write to Rutherford instead.

Rutherford replied:

June 25, 1910

Dear Sir,
I have received your letter asking permission to work at the Physical Laboratory of the University Manchester on radioactivity. I shall be pleased to consider favorably your proposal provided you are able to spend at least a University year in the Laboratory. Some little time is required to become acquainted with the methods of measurement and it is not possible to hope to accomplish even short research under that period. I may mention for your guidance that you could probably be admitted as a “Research student”. This entails a fee of 9-9-0 per year to the University and covers the greater part of the University expenses. The Laboratory provides all the apparatus and facilities for research free of charge. I shall be glad to hear from you whether you will be able to come under these conditions.

Yours sincerely E. Rutherford
Hevesy in Manchester

So on January 19, 1911 he had started his ‘training’ in radioactivity, under the guidance of Rutherford’s assistant Geiger, by counting α-rays. On the very next day he isolated the decay products of actinium-emanation and determined their half-lives. January 21, 1911 he started the measurement of the recoil of atoms after alpha emission. [Hevesy G 1911] Following on from these successes Rutherford suggested Hevesy investigate the solubility of actinium-emanation in water. This was a complicated task, because its half-life amounts to only 3 seconds. He used a flow-through technique, Charles Darwin helping him developing the mathematical theory. [Hevesy G 1913] On a personal note he occasionally enjoyed dining at the Midland Hotel with Geiger and Darwin. [Cockroft 1967 b]

Hevesy’s interest in x-ray spectroscopy

Hevesy and Moseley became close friends. Hevesy selected a suitable crystal of potassium ferrocyanide for the use in X-ray spectroscopy, and managed to secure a very rare sample of metallic tantalum which he obtained from his friend Werner von Bolton, who had first prepared metallic tantalum. With Moseley he had a lot of fruitful discussions about the possibility of separation of radio elements with the same chemical behaviour but different atomic masses and made much use of X-ray spectroscopy in the search for unknown chemical elements. During ‘tea time’ in Rutherford’s institute in 1913 he commented to Moseley that it would be interesting to trace the way the components of tea passed through the human body. This was a hint of Hevesy’s thinking which led to important discoveries in the second half of his scientific career. Since that time he was always looking for the possibility of applying the indicator method in biology and medicine.

His friendship with Moseley gave him the opportunity to assist in the construction of his spectrometer. He was much impressed by Moseley’s invention of the law of correlation between X-ray energy and the atomic number of the target element used and saw the possibility of using this method to search for undiscovered elements and the determination of elemental concentrations. He wanted to work with him in the summer 1914 but before Hevesy could travel to England, World War I had begun and Moseley joined up. He died in a battle in the Dardanelles. Hevesy later used X-ray spectroscopy to discover the element hafnium and to develop and use x-ray fluorescence analysis to determine trace elements in rocks.
Physicochemical investigation of radio elements

In Manchester his interest changed drastically. The main questions being discussed in the Rutherford group concerned the placing of the large number of newly discovered radioactive elements in the periodic table, the structure of the atoms, and the source of radioactivity. Hevesy studied the valences of the radioactive elements by means of physicochemical methods [Hevesy G 1912a; 1913a; 1913b]xiv, xv, xvi. According to Soddy’s α-ray rule by which, after the emission of an α-particle the remaining atom becomes two places lower in the natural sequence of elements. Hevesy would demonstrate this by diffusion measurements.

In the winter of 1912 during a stay in the sanatorium Mariagrün near Graz (Austria) he studied tables of physico-chemical data, where he found interesting examples of the relation between ion radii and the diffusion coefficient. After Hevesy had characterized the radioactive members of the decay chain by its electrochemical potential he measured their diffusion coefficient from which he obtained their valence and its change after α – and β – decay. He had not expressed directly the corresponding changes of the atomic number in the periodic table of the elements according to the ideas of Frederick Soddy and Alexander Fleck in Glasgow and A. S. Russell from Manchester. Meanwhile Fleck at Soddy’s suggestion applied the co-precipitation method to verify the valence of the daughter nuclide after decay. In autumn 1912, Russell reported about a short visit in Glasgow that Fleck had extended Soddy’s rule to the β -decay. When Hevesy planned to spend the Christmas of 1912 in Budapest with his family, Kasimir Fajans, who discovered, in 1911, with Makower in Manchester, the first branching in a radioactive decay series, [Fajans K 1911]xvii invited him to stop over in Karlsruhe. In 1912 Fajans had obtained the position of assistant in the Institute of Physical Chemistry in Karlsruhe and was much interested in the results of Hevesy and Fleck. After the discussion with Hevesy, Fajans intended to write a paper about the decay rules and being concerned about the work of Fleck and Russell Hevesy proposed to Russell that he should write a paper as soon as possible.

Discussion with Niels Bohr about the atomic model

In March 1912 Niels Bohr arrived in the laboratory and on the 16th of that month started working in radioactivity - and like all newcomers before, he had done some measurements with α-rays. When he arrived, Rutherford
was actually on holiday but during teatime, Geiger, Marsden, and Makower told him about the new atomic model of Rutherford. Bohr immediately began considering the problem that electrons circulating around the nucleus lost their energy because of the electrical rules. Over a long time, Hevesy discussed with him daily on the structure of atoms, the reason why in Rutherford’s model, the electrons did not fall into the positive nucleus and the reasons underlying the periodic table. Hevesy’s knowledge about the chemical behaviour of the elements was very helpful for Bohr in the development of his atomic model and a lifelong friendship with him began. In 1919 and 1934 when Hevesy lost his positions in Budapest and Freiburg, respectively, Bohr offered him an opportunity to work in his institute in Copenhagen.

Unsuccessful experiments of separation RaD from lead

The most important invention of Hevesy is the use of isotopes as indicators in the study of chemical processes. (The Nobel Prize in Chemistry, 1943, was awarded to George de Hevesy "for his work on the use of isotopes as tracers in the study of chemical processes". Hevesy used the term indicator, the term tracer was used later - the title of his Nobel Lecture on December 12, 1944, was “Some applications of isotopic indicators”)

Some of the discovered radioactive elements seemed to be chemically similar and inseparable. The term "isotope" had not yet been coined. One day in autumn 1911 he met Rutherford in the cellar of the institute where some hundreds of kilograms of lead chloride, presented by the Austrian Government, were stored. It was obtained after extraction of uranium and radium from Jachymov pitchblende. It contained large amounts of the valuable radioactive RaD, now known as $^{210}\text{Pb}$. Rutherford, characteristically, said to Hevesy:

"My boy if you are worth your salt you will separate the RaD from all that nuisance of lead". [Hevesy G 1962] xviii #

Recalling later, Hevesy wrote, “in those days” I “was an enthusiastic young man and embarking soon on the separation experiment, thought he could succeed.” At that time Rutherford and Hevesy did not imagine, that RaD was an isotope of lead ($^{210}\text{Pb}$) - which could not be separated with chemical methods then available in the laboratory.
The later use of RaD as the first radioactive indicator was very important. In 1898 Marie and Pierre Curie were searching for the unknown radioactive element responsible for the higher radioactivity of pitchblende than was accountable by the uranium. \textsuperscript{xix} They found no radioactivity measured by air ionization\textsuperscript{xx} in lead separated from pitchblende \textsuperscript{xxi} because of the ionization in air by the $\alpha$-rays of $^{210}$Po is about two orders of magnitude higher than by the $\beta$-rays of $^{10}$Pb and $^{210}$Bi.

In 1900, in Munich, the chemist Karl Andreas Hofmann \textsuperscript{xxii} had measured, together with his student Eduard Strauss, chemically isolated substances in uranium and rare earth minerals \textsuperscript{xxiii} by the darkening of photo plates, and found that lead separated from uranium and thorium containing minerals were radioactive. After separation of bismuth from lead, they found that the lead remained radioactive and suggested that they had discovered a new radioactive element which was similar to lead and named it radio-lead. \textsuperscript{xxiv}

In 1902 Rutherford found that the radioactivity, which was collected on electrodes in the vicinity of radium, was growing and decaying with time after the removal of the radium. He assumed that the radioactive substance at the electrodes emitted two kinds of radiation. He found that this radioactivity could be dissolved from the electrodes by dilute acids.\textsuperscript{xxv}

Interpreting the decay curve he identified three radioactive substances which he named radium A, radium B and radium C.

In 1904 he was looking for slow decaying decay products of the emanation. After he termed the first new product Radium D which half transformed in about 40 years and wrote:

"It is now necessary to consider the question whether the substances radium D and E have been previously separated from pitchblende, and are known by other names. In regard to radium D, there is some doubt whether it has been previously separated. It is possible that it is the radioactive constituent present in the radio-lead of Hofmann, for he states that this substance emits a large amount of $\beta$ - rays."\textsuperscript{xxvi}

At that time nobody understood that the same element isolated from different minerals could be in one case radioactive and in another not. Therefore it was assumed that lead and radio-lead are different chemical elements. Rutherford wrote in his book about radioactivity which was published in 1913 that
“it would be desirable to extract pure radium D from pitchblende. After one month its beta activity would be 30 times more than the activity of same weight of radium.” xxvii

In 1908 the Hungarian, Bela Szilard, who spent some time in the laboratory of Marie Curie, tried to separate radio lead from lead. In one of his experiments he found that radio-lead was separated from lead.xxviii Two years later Heinrich Herchfinkel from Switzerland could not separate these two elements. xxix

Because of the fact that RaD ($^{210}$Pb) has only low energy β-rays to measure, Hevesy had to wait some weeks until enough disintegration product RaE ($^{210}$Bi) had built up. In some of his first experiments in the autumn of 1911, he had not waited long enough and although the separation of RaD and lead seemed to be successful but in fact it only resulted in the separation of lead and bismuth. At the end of 1911 Hevesy became depressed because he had been unsuccessful in the separation of RaD and lead. On the way to Budapest, in December, he visited the new Radium Institute in Vienna, which had been founded in 1910. About this he wrote to Rutherford, saying that in the laboratory in Vienna a man was working on the same problem. Hevesy had not told the head of the Institute, Prof. Stefan Meyer, that they had tried to separate RaD and lead, too [Hevesy G 1912 D]. About the institute in Vienna he wrote that “they have many rooms, new instruments, and much radium”.

After his stay in Budapest and in the sanatorium near Graz he spent some weeks with his family in Lovrana (now Croatia) on the coast of the Adriatic Sea, took part at the meeting of the Bunsen Society in Heidelberg, and came back to Manchester at the end of the spring of 1912. Meantime Rutherford had become rather impatient over Hevesy’s long absence and asked another member of his group, J. N. Pring, to continue the separation experiments of RaD and lead. Hevesy maintained correspondence with him (although none has been yet discovered), and hoped that he would be successful in the enrichment of RaD. [Hevesy G 1912 E]xxx. After returning to Manchester in the summer of 1912 he systematically studied the possibilities of the separation of elements which are chemically identical but different in mass. In the following year these were identified by Frederick Soddy as isotopes by physical-chemical methods mentioned in the chapter above, and discussed this problem with Moseley and Bohr.
After lengthy trials Hevesy arrived at the depressing result that separation of RaD from lead could not be achieved. But because RaD could not be separated from lead Hevesy had the idea to take advantage of this and to use chemically inseparable radioelements as a tracer for stable one. He thought that it should be possible to add RaD prepared free of lead from old radium emanation tubes to a small known amount of lead and thus label the lead.  

First he wanted to determine the very low solubility of lead chromate in water using RaD as an indicator for lead. Travelling between Manchester and Budapest he again made a visit to the Radium Institute in Vienna and there he learned that Friedrich Paneth also had had no success in separating RaD and lead. Spending Christmas in Budapest Hevesy wrote to Paneth in the last days of 1912:

“I want to propose you: Because lead and radium D are not separable, we should take RaD as indicator of lead, and for instance investigate the solubility of PbCrO₄ in water at different temperatures”.

He proposed that they work on this idea together and thus they demonstrated the use of radioactive isotopes as tracers. It was done in the Radium Institute, because of the better facilities for chemical experiments and they successfully published the first application of a radioactive indicator. They also published the unsuccessful experiments to separate RaD and lead.

The indicator method became an important tool in a large number of different fields in science, technique and medicine in the second half of the last century and is now successfully applied in medical diagnostics. Hevesy had used this method for the investigation of transportation, distribution and reaction of elements and compounds in physical chemistry, physiology and medicine and therefore he was awarded the Nobel Prize in Chemistry in 1943. For the importance of his work in the medical diagnostics he was named “father of nuclear medicine”.

**Hevesy’s discoveries based on his research and discussions in Manchester**

*Radioactive tracer for physicochemical investigations.*

In the winter of 1913 Hevesy got his lecturing qualification at the University of Budapest. Travelling between Manchester, Vienna and Budapest, he applied the indicator method in Physical Chemistry using
RaD, E ($^{210}$Bi) and F ($^{210}$Po). Hevesy solved a number of fundamental questions in physical chemistry, e.g. the self-diffusion of the atoms in molten and solid lead xxxvi and the velocity of the exchange of atoms between different ionic and molecular compounds of lead. As a Hungarian, in 1916 Hevesy had to serve in the army. After the war in 1918 he stood in for the ailing professor of physics at the University of Budapest and in 1919 while the communist Bela Kun was leader of the Council Republic, he was appointed to the Chair of physical chemistry at the Budapest University. After some month Romanian troops invaded the country, and Admiral Horthy came to power. Hevesy lost his chair and the allowance of teaching at a Hungarian university xxxvii. He worked some months on the self diffusion of molten lead with Gyula Groh in his chemical laboratory of the veterinary college and there he also studied the exchange of $^{210}$Pb in inorganic and organic compounds of lead with Laslo Zechmeister. xxxviii With these results he confirmed the theory of ions in solutions of salts, which had been established by Svante Arrhenius. In 1920 he emigrated to Denmark. There Hevesy could continue experiments with radioactive isotopes and he studied the transfer of lead using $^{210}$Pb from the soil in different parts of bean plants. xxxix

Separation of isotopes

In Copenhagen his friend Niels Bohr found a place for him in the laboratory of Johannes Nikolaus Brönsted (1879-1947) which provided a base for him. During his stay in Manchester Hevesy had had some ideas to separate isotopes by physical methods, taking advantage of their different masses, and separated isotopes of mercury and chlorine xl and later of potassium by distillation.

Discovery of hafnium

In Manchester, Bohr and Hevesy came to the conclusion that the atomic number and not the atomic weight are responsible for the chemical properties of an element and its place in the periodic table. Moseley, who discovered the dependence of the energy of X-rays on the atomic number, verified this idea. In Copenhagen Bohr was interested in the X-ray spectra of different elements and invited Dirk Coster to install a spectrometer in his institute. Hevesy assumed that the missing element 72 should be a homologue of zirconium and planned the search of this element in zircon minerals. In 1922 Coster and Hevesy analysed zircon minerals by X-ray spectroscopy, and found the line of the then unknown element number 72
which they named hafnium\textsuperscript{xli, xlii}. Hevesy enriched hafnium by fractionated crystallisation of potassium zirconium fluoride and they determined the abundance of Hafnium in different minerals and zirconium compounds.

The development of X-ray fluorescence analysis

After the discovery of hafnium he received offers from German universities and in 1926 he took a chair at the University in Freiburg. He became director of the Institute of Physical Chemistry and despite his Hungarian citizenship, citizen of the state of Baden in Germany. There he applied the X-ray spectroscopy techniques for the determination of trace elements in minerals and rocks and developed together with his assistants and students the X-ray fluorescence analysis. He was interested in elemental abundances, because in his opinion the abundances are determined by the stability of their nuclei, which are built in the stars by nuclear reactions\textsuperscript{xliii}.

Further discoveries in Freiburg

Hevesy was interested in geo- and cosmo- chemistry. He was in close contact with the founder of modern Geochemistry Viktor Moritz Goldschmidt, whom he had met when both were students in Freiburg. Hevesy was also in contact with the Geochemist Arthur Holmes from Durham who had determined the age of rocks using the relation of the content of uranium and lead formed by the decay of uranium in uranium minerals. Hevesy had some doubts that the determinations of lead were precise so he added a known amount of radioactive lead to a mineral before separation and after the separation of lead he measured the activity of the obtained lead. The loss of radioactivity was proportional to the loss of separated lead. This method is named isotope dilution analysis and is used in radiochemical analysis and in inductive coupled plasma mass spectrometry.

Hevesy assumed that beside, uranium, thorium, their decay products, and potassium further radioactive elements may exist. With help of a sensitive counter obtained from his friend Hans Geiger he looked for radioactive rare earth elements (r. e. e.). He identified in some of the r. e. e. the measured radioactivity as impurities of known radioactive isotopes. In samarium the radioactivity remained after further separation experiments. Thus he discovered the radioactivity of samarium. (His search for a stable isotope of polonium by X-ray fluorescence analysis was not successful).

When his friend Urey has discovered heavy hydrogen, he sent a few liters of labeled water containing 0.6% of D\textsubscript{2}O to Hevesy, who used it
immediately for the investigation of hydrogen exchange in a fish and his own body. This was the first use of a stable isotope as indicator. After the national socialists in Germany came to power, he decided to go again to Copenhagen.

**Neutron activation and application of radioisotopes in physiology in Copenhagen 1934 - 1943**

At the time when he left Germany artificial radioactivity had been discovered and small neutron sources were available. In Copenhagen Hevesy first used the pure r. e. e. he obtained from Auer v. Welsbach and hafnium to study the radioisotopes formed by neutron activation. Together with Hilde Levi, he observed the extremely high activation yield of dysprosium and europium, which gave him the idea of taking advantage of this fact by a new analytical method, the neutron activation analysis.

Hevesy applied this new method for purified r. e. e. and studied details, which were important for the application of this method.

**Radionuclides in biochemistry**

Through the years he was disappointed that biochemical processes could not be investigated with the help of the known radioisotopes because of the fact that relevant biological elements were stable. The situation changed drastically after the discovery of artificial radioactivity. Then he was able to use $^{32}$P and later, other radioactive isotopes as indicators in life sciences. His first experiments with this nuclide were devoted to the metabolism of phosphorous in solid organs like bones and teeth. He had used a Rn-Be neutron source for the irradiation of carbon disulphide to produce $^{32}$P. With this isotope he studied the distribution and kinetic of exchange of phosphorus in different parts of animals, beginning with teeth and bones. For this work he first co-operated with the surgeon of the Finsen Hospital, Ole Chievitz, a friend of Niels Bohr, getting the surprising result of the continuous exchange of phosphorus in the bones. When they published the results the biologists would not believe them. Then he was working with many scientists in different laboratories in the field of nuclear physiology.

In 1943 when it became dangerous to stay in Denmark he decided to flee to Stockholm where he continued his work with radioactive tracers in biology, physiology and medicine in the Institute of Organic Chemistry headed by Hans von Euler-Chelpin. In 1944 he got the Nobel Prize for Chemistry for the year 1943 for founding the indicator method and its application in biology and medicine. After WWII he decided to choose Sweden for his
permanent residence and used the possibility to obtain Swedish citizenship which was available for Nobel Prize laureates. In Stockholm Hevesy was working on different problems in laboratories of different institutes and cooperated with a great number of scientists.

All his work about life science he summarized under the chapters: Skeleton – Phosphatides – Permeability – Labelled Blood Corpuscles in his collected publications.

Hevesy was acquainted with a lot of famous scientists of different disciplines all over the world. He was travelling all his life, at the beginning to learn as much as possible, later, in order to find interesting places for research and to secure his own life, to present his experiences by lectures, to receive awards, honorary degrees and memberships in learned societies, to visit friends and to find places for his health. In 1949 he was decorated with the Copley medal of the Royal Society. He asserted that for him this award was the most important one.

From 1949 he visited Freiburg each year for some weeks at a time. He had loved the city since his time as student and as professor, and had many friends and colleagues there. He became Dr. h. c. nat. in 1949, and in 1959 Dr. h. c. med. From 1952 he took part in the annual meetings of the Nobel Prize Laureates in Lindau at the Lake Constance where he give lectures about his physical, chemical, and physiological research. In a new building of the Radiological Institute he had his own room for his studies. His main interest was cancer research and hematology. Whilst in the medical clinic of the university, the use of radioisotopes for research and diagnostics was begun and he arranged the supply of the isotopes from his friend Ernest O. Lawrence in Berkeley (California).

In 1962 when his health was deteriorating, he consulted Ludwig Heilmeyer, the head of the medical clinic whom he knew from his hematological research. Heilmeyer diagnosed a lung cancer, and offered him treatment in his clinic. Sadly, and ultimately to no avail, as he died there on July 6, 1966, and was buried in Freiburg - Littenweiler.

J. D. Cockcroft wrote the Biographical Memoires about him as a Fellow of the Royal Society basing it on a manuscript written by Hevesy himself.

After his death he was more and more forgotten in Freiburg. When his daughter Ingrid, who lived in Freiburg, no longer felt able to take care of
the grave, the Hungarian Academy of Sciences came to their aid and arranged a reburial, in Budapest on April 19, 2001 at the Kerepesi cemetery in the centre of the town of his birth, attended by his children and grandchildren.

In conclusion

After Hevesy became familiar with the counting technique of radioactive samples, he noted the different behaviour in decay but similar chemical behaviour of the emanations of uranium and actinium, as an example of the, later identified, isotopes. When Hevesy was unable to perform Rutherford’s second task, i.e. the separation of radium D and lead, he had the idea of using radium D as an indicator for lead and thus discovered the ‘indicator method’, which he applied successfully in chemistry, geology, physiology and medicine. In Manchester he had learned x-ray spectroscopy techniques from Moseley. Using this method he discovered the element hafnium and used this principle for the analytical determination of trace elements and the development of the x-ray fluorescence method (XRF). Also in Manchester he had met Niels Bohr, with whom he discussed the atomic model, and in the laboratory he made a lot of friends which opened the door to the British scientific community.

There is little doubt that Hevesy’s successful scientific career was profoundly influenced by his time working with Ernest Rutherford in Manchester

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