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Nobel Prize awards in Radiochemistry

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Dedicated to the memory of late Karl H. Lieser, Gerhard L. Stöcklin and Alfred P. Wolf with whom the author shared the editorial work of Radiochimica Acta from 1977 to 1995

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Summary. In 1996 the Editors of *Radiochimica Acta* brought out a special volume of the journal to celebrate the hundredth anniversary of the discovery of radioactivity [1]. On the occasion of the 50th anniversary of *Radiochimica Acta*, which follows closely upon the centenary of Marie Curie's second Nobel Prize in 1911, the author has the privilege to informally review "Radiochemistry and Nobel Prize Awards", including discoveries of radioelements and new fields in chemistry based on radiochemical methods.

1. The beginning

The Nobel Prizes in Physics and Chemistry were established in 1901, six years after the discovery of radioactivity and three years after the discoveries of the elements polonium and radium. They are awarded by *Kungliga Vetenskapakademien* (the Royal Swedish Academy of Sciences) on the basis of proposals made by respective Committees on Physics and Chemistry, which receive recommendations from Swedish and foreign scientists [2]. The first Nobel Prize in Physics was awarded to Wilhelm Conrad Röntgen (1845–1923) "in recognition of the extraordinary service he has rendered by the discovery of the remarkable rays subsequently named after him".

On March 2, 1896 Antoine Henri Becquerel (1852–1908) (Fig. 1), grandson, son and father of eminent scientists, while pursuing experiments driven by Röntgen's observations, found a different kind of radiation emitted by uranium compounds. Within several months Becquerel published in the *Comptes Rendus* of the *Académie des Sciences* about ten short articles on "invisible radiations emitted by uranium" [3, 4]. However, interest in the new field faded rapidly. X-rays were the subject of a thousand papers and 50 books in 1896 alone, while reports on uranic rays did not exceed about thirty by the end of 1897. Becquerel himself became more interested in the Zeeman effect. Lawrence Badash (1934–2010) commented, "Becquerel should not be criticized overly for departing from his study of uranium

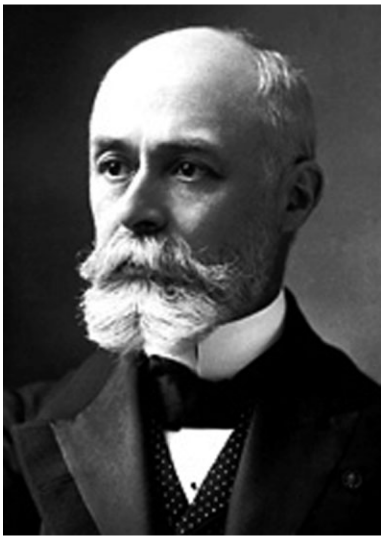

Fig. 1. Antoine Henri Becquerel (1852–1908).

rays when he thought the subject was exhausted. By the end of 1897 radioactivity was something of a dead horse: it was there, but no one knew what to do with it" [5].

At that time Pierre Curie (1859–1906) and Marie Curie (1867–1934), née Maria Salomea Skłodowska, (Fig. 2) came upon the scene. Within a few months, from April to December 1898, they discovered two new elements, polonium and radium, that they identified solely by the emission of Becquerel radiations [6, 7]. In their first publication (April 22, 1898) in the weekly *Comptes rendus* of the Académie des Sciences Marie Curie reported that certain uranium-bearing minerals, in particular pitchblende, emitted more rays than metallic uranium and concluded, "this fact is quite remarkable and suggests that these minerals may contain an element much more active than uranium itself" [8]. Three months later, in an article of July 18, 1898, "On a new substance, radio-active, contained in pitchblende" [9], the Curies (with Pierre as senior author) confirmed their supposition. The title contained the first mention of the word *radio-active*. The Curies omitted the hyphen in the following year.

Spectroscopy did not reveal any characteristic line of the new element, and the authors cautiously suggested, "if the

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Fig. 2. Pierre Curie (1859–1906) and Marie Curie (1867–1934).

existence of this new metal is confirmed, we propose that it be named polonium in honour of the native land of one of us" [10]. On December 26, 1898 the Curies announced the discovery of a second, strongly radioactive substance similar to barium. The activity in the final batch was 900 times greater than that of uranium, and the spectroscopic analysis revealed lines that could not be assigned to any known element. The Curies concluded, "we think that the new radioactive substance contains a new element to which we propose to give the name radium" [11].

2. The expansion of radiochemistry

The discoveries of polonium and radium in 1898 were the landmarks of research in the new sciences of radioactivity and radiochemistry. Becquerel returned to the field as early as 1899. André Debierne (1874–1949) joined the Curies and in 1899 discovered a third radioactive element, actinium (atomic number 89) [12]. Julius Elster (1854–1920) and Hans Geitel (1855–1923) were the first scientists in Germany to become interested in Becquerel radiation and radioactivity [13]. The German Friedrich Oskar Giesel (1852–1927) and the Austrians Stefan Meyer (1872–1949) and Egon von Schweidler (1873–1948) undertook a large-scale separation of radium and polonium from residues of uranium ores [14]. In 1898 Ernest Rutherford found that uranic rays had two components, which he named the α- and β-rays for "convenience". In the United States the Becquerel experiments were reviewed as early as April 1898. The discovery of radium was announced in the January 28, 1899 issue of *Scientific American*, scarcely one month after the publication in the *Comptes Rendus*. The annual entries in the bibliography on radioactivity increased steadily (except for a drop during World War I and the next few years) with peaks around 1908 (the year of Rutherford's Nobel Prize), 1934 (the Joliots' announcement of artificial radioactivity) and 1939 (Hahn and Strassmann's discovery of nuclear fission) with an estimated 4000 publications on the outbreak of World War II.

3. The crossed itineraries of Nobel laureates in radiochemistry

Marie Curie and the Joliot couple are an exception among Nobel laureates: they performed their whole scientific work in their Parisian laboratories. The Curies and Ernest Rutherford, the most famous figures in the early history of radiochemistry, met once in Paris, on June 12, 1903, the day of the presentation of Marie Curie's thesis at the Sorbonne University. Rutherford remembered the evening party shared with Paul Langevin (1872–1946) and Jean Perrin (1870–1942; Physics Nobel laureate 1926):

"After a very lively evening Professor Curie brought out a tube connected in part with zinc sulphide and containing a large quantity of radium in solution. The luminosity was brilliant in the darkness and it was a splendid finale to an unforgettable day. At that time we could not help observing that the hands of Pierre Curie were in a very inflamed and painful state due to exposure to radium. This was the first and last occasion I saw Curie. His premature death in a street accident in 1906 was a great loss to science and particularly to the then rapidly developing science of radioactivity" [15].

It is amazing how fast the Curies' pioneering work spread among laboratories in France, Germany, Austria, UK, Canada and USA. Researchers in the field became acquainted and moved to several laboratories before settling in definitive positions. The leading personality in the new science of radioactivity was undoubtedly Ernest Rutherford whose prodigious work attracted further researchers in the field including later Nobel winners Hahn, Soddy and Hevesy.

The Nobel Prize awards in radiochemistry cover a timescale of about 40 years, from the discoveries of polonium and radium to the synthesis of the first transuranium elements.

4. 1903 – Henri Becquerel, Pierre Curie and Marie Curie

Radioactivity was relevant to physics, whereas the discovery of the new elements belonged to chemistry. Initially, the dilemma did not seem to raise any serious problem. Essentially, uranic rays were at the origin of the search for new elements. Thus nominees could be presented to the Committee for Physics. Nevertheless, in the history of Nobel Prizes, awards for the discovery of elements remained the prerogative of the Committee for Chemistry [16–18].

Becquerel and Curies were first nominated for the Physics Prize in 1902. June 13, 1903 Marie Curie defended at the Sorbonne her thesis "Researches on radioactive substances". The board of examiners included two Academicians and future Nobel laureates, Gabriel Lippmann (1845–1921) and Henri Moissan (1852–1907). These eminent scientists considered Marie Curie's discoveries as the most important results ever presented in a doctoral thesis. When Pierre Curie learned that he was sole nominee with Becquerel, he insisted that Marie's initial work led to the discovery of the new elements and "many people would be surprised if we were separated in this circumstance".

In June 1903, before the Swedish Academy had made the final decision, the Curies were invited by the prestigious Royal Institution in London to present a Faraday lecture to an audience of the major scientists in the country. November 1903 they were awarded the Humphry Davy Medal, given each year "for the most important discovery in chemistry". These successive honours culminated on November 12, 1903 with the decision of the Swedish Academy to assign the Nobel Prize to Becquerel and the Curies: "One half to Henri Becquerel in recognition of the extraordinary services he has rendered by his discovery of spontaneous radioactivity, and the other half jointly to Pierre Curie and Marie Curie, née Skłodowska for their joint research on the radiation phenomena discovered by Henri Becquerel".

After formally addressing his thanks to the Academy, Pierre Curie declared straightaway that neither he nor his wife would attend the ceremony owing to lecturing responsibilities and Marie's illness. December 10, 1903, Becquerel presented in Stockholm his lecture "On radioactivity, a new property of matter". He made an honest attempt to recapitulate the Curie's work. Eventually Pierre and Marie Curie travelled to Sweden in June 1905, 18 months after the award. In name of the couple Pierre Curie presented a short Nobel lecture, "Radioactive substances, especially radium", beginning "we tender you our apologies for being so tardy in visiting you, for reasons outside our control".

5. 1904 – Sir William Ramsay

Sir William Ramsay (1852–1916) (Fig. 3) was involved in the Nobel Prize for Chemistry as soon as 1901 [19]. He was one of the 11 chemists who nominated the first laureate, Jacobus Henricus van't Hoff (1852–1911). Thereafter, he was a nominee each year from 1902 to 1904 when he won the prize with 23 nominations, the largest number of nominations received for a single candidate up to that time, including those of Nobel laureates Emil Fischer and Van't Hoff, for "the discovery of the inert gaseous elements in air and the determination of their place in the periodic system". Ramsay was the first Briton to receive the Chemistry Prize.

The same year, the Nobel Prize in Physics was awarded to the physicist Lord Rayleigh (1842–1919), 3rd Baron Rayleigh, born John William Strutt, "for his investigations of the densities of the most important gases and for his discovery of argon in connection with these studies". The two Nobel laureates worked together and on August 13, 1894 they announced jointly the discovery of a new element, the first inert gas, which they called "argon" [20].

Ramsay pursued the search of gases in air and by 1900 he had added helium, neon, krypton and xenon to argon. In the meanwhile Rutherford observed that a radioactive material was emitted by thorium and the German chemist Dorn (1848–1916) discovered a similar emission from radium which he called "emanation". In 1904 Rutherford introduced the name "radium emanation" which was eventually changed to "radon".

Ramsay had a strong proprietary feeling of noble gases and was anxious to make his own investigation of the properties of the new radioactive gas. Incidentally, in 1903

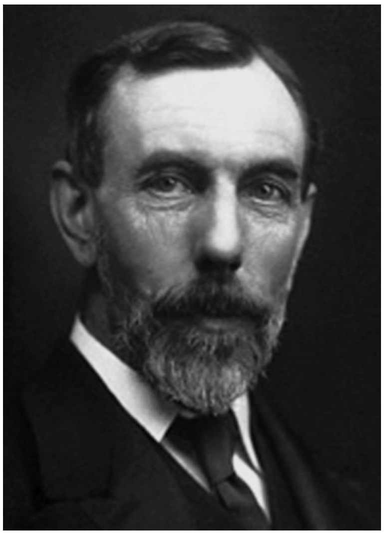

Fig. 3. William Ramsay (1852–1916).

Frederick Soddy moved from Rutherford's laboratory in McGill (Montreal, Quebec) to William Ramsay's at the London University College where he spent one year. Together, the chemist and the expert in the isolation of minute quantities of gases examined the spectrum of the "radioactive emanation" of 20 mg of radium bromide and confirmed that it was identical to the spectrum of helium [21]. This observation confirmed Rutherford's disintegration theory and that radium was an element as Marie Curie had already proved. Although the Ramsay-Soddy discovery was a landmark in the history of radioactivity it would not alone deserve a Nobel award.

Ramsay, without particular experience in the field, pursued researches on radioactivity of his own and claimed that in addition to helium, neon and argon were also emitted by radium or produced by radon on contact with water and furthermore that radon would transmute copper into lithium or sodium. In a letter to his friend the American chemist and physicist Bertrand Borden Boltwood (1870–1927), Rutherford noted "If Ramsay is right, the subject of radioactivity enters a new phase ... he has a lot of enemies who do not believe a word of what he says. The weaknesses of his results lie in the fact that the neon result has only been got once; the radon is frisky while the lithium I must confess is more than I can momentarily swallow. Unfortunately nobody has any radium to test his results ..." [22]. Unlike Rutherford, Marie Curie did have enough radium to verify Ramsay's results about copper being transmuted to lithium. Together with the Norwegian Ellen Gleditsch (1879–1968) she repeated Ramsay's experiment and proved that the traces of lithium reported by Ramsay emanated from the glass vessel used in the experiment. When pure platinum containers were used to perform the same experiment, lithium was no longer detected [23]. August 23, 1908 the New York Times reported "Metals not transmuted: Mme Curie finds no proof that radium makes sodium from copper salts".

Soddy, in an obituary of his former mentor in London, said "Pre-eminent in chemical science before the discovery of radioactivity, Ramsay devoted the last ten or twelve years of his vigorous and crowded life more and more exclusively to the young science he did so much to advance".

6. 1908 – Ernest Rutherford

In 1907, Svante Arrhenius nominated Rutherford (1871–1937) (Fig. 4) to the Nobel Prize in Chemistry. The same year Rutherford had seven nominations to the Physics Prize, including those from Nobel laureates Emil Fischer (Chemistry, 1902); Philipp Lenard (Physics, 1905); Adolph von Baeyer (Chemistry, 1905), and the to-be (Physics 1918) laureate Max Planck.

The Chemistry Committee reported to the Academy that the single nomination in Chemistry is "understandable, taking into account that Rutherford uses physical methods while the results, so far as they are concerned with chemical elements, must be considered to be of fundamental importance also for chemistry". The proposal of the Committee was acknowledged by the Academy:

"Though Rutherford's work has been carried out by a physicist and with the aid of physical methods, its importance for chemical investigation is so far-reaching, that the Royal Academy of Sciences has not hesitated to award to its progenitor the Nobel Prize designed for original work in the domain of chemistry – thus affording a new proof to be added to the numerous existing ones, of the intimate interplay one upon another of the various branches of natural science in modern time".

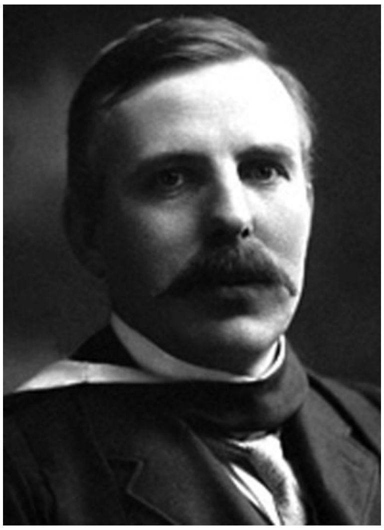

Fig. 4. Lord Rutherford of Nelson (1871–1937).

In 1908, Arrhenius nominated again Rutherford for the Chemistry Prize. He was joined by the Swedish Oskar Widman, member of the Nobel Committee for Chemistry and the Austrian Rudolf Wegscheider. To ensure Rutherford's award of a Prize, Arrhenius nominated him also for the Physics Prize: "If the Academy should decide that it is not appropriate to give him the Chemistry Prize, he should be awarded the 1908 Physics Prize". Eventually in 1908 Rutherford had seven nominations in Physics and three in chemistry and was nevertheless winner in Chemistry "for his investigations into the disintegration of the elements, and the chemistry of radioactive substances".

At the banquet speech following the award ceremony of the 1908 Nobel Prize in chemistry, Ernest Rutherford said "I have dealt with many different transformations with various periods of time, but the quickest that I have met was my own transformation in one moment from a physicist to a chemist" [24]. Among the headings of Rutherford's around two hundred publications, only one refers to chemistry, namely his Nobel lecture "The Chemical Nature of the Alpha Particles from Radioactive Substances".

After a short term from 1895 to 1898 at the Cavendish Laboratory in Cambridge (UK) where his talents were recognized by the 1906 Physics Nobel laureate Joseph John Thomson (1856–1940), discoverer of the electron, Rutherford moved to the McGill University in Montreal, Canada where he started his research on radioactivity and published about 70 articles. His first work dealt with the emanation of a radioactive gas (thoron) from thorium compounds [25] and the radioactivity induced by the action of thorium [26]. In collaboration with Soddy, Rutherford published in 1902 the first comprehensive review in the field initiated by the Curie: "The cause and nature of radioactivity" [27]. In 1904 he received the Rumford Medal, his first major scientific Prize.

When he moved to Manchester in 1907 he had published over 70 articles in the field of radioactivity. He returned to Cambridge in 1919 where he pursued his prodigious scientific work.

As a Nobel laureate, Rutherford was invited to nominate Nobel Prize candidates. Among his eight nominees in Physics, seven became Nobel laureates, including Niels Bohr (1922), and James Chadwick (1935). In 1935, Rutherford nominated also Frédéric Joliot and Irène Curie to the Chemistry Prize.

Rutherford received the Prize in Chemistry although his work dealt essentially with Physics. It could be expected that a second Prize for Physics may complete the first Nobel award. He was nominated for the Physics Prize by Theodore Svedberg in 1921 and 1922, David J. S. Jordan in 1924 and Johannes Stark five times between 1931 and 1937 [24]. These nominations were discarded by the Swedish Academy on various grounds. The argument of two Nobel Prizes awarded to Marie Curie was refuted because she had shared her first Prize with Pierre Curie and Becquerel. Nevertheless, Rutherford is the only Nobel laureate to have performed his most famous discoveries *after* receiving the Prize: the atomic nucleus, his greatest contribution to Physics in 1911, and the first transmutation in 1917, converting nitrogen to oxygen. Rutherford pursued research until his death, October 19, 1937. His last publication August 21, 1937 was "Search for the isotopes of hydrogen and helium of mass 3". His ashes were buried in the nave of Westminster Abbey, next to the tombs of Sir Isaac Newton and Lord Kelvin.

After a dispute between Russian and American groups, IUPAC decided in 1997 that the element with atomic number 104 would be rutherfordium, Rf.

7. 1911 – Marie Curie's Nobel Prize in Chemistry

In the history of Nobel Prizes, Marie Curie is the first woman laureate, the first recipient of two awards and still the only laureate in Physics and in Chemistry [28, 29] (Fig. 5). After Pierre Curie's tragic death on January 19, 1906, the Faculty of Sciences entrusted Marie Curie with the direction of the laboratory. Pierre's Chair of Physics at the Sorbonne and his seat at the Académie des Sciences were now vacant. The academic authorities appointed quickly Marie the first woman professor at the Sorbonne. Her inaugural lecture on November 5, 1906 was celebrated as a victory for feminism. In November 1910, to increase the chances of obtaining funds for her laboratory, Marie Curie offered herself as a candidate for a vacant seat at the *Académie des Sciences*, an exclusively male body. Although she failed to be elected because of misogyny, she was the first woman elected an Associate member of the *Académie de Médecine* for "her contribution to the discovery of radium and a new treatment, the Curie therapy".

However, there were still doubts about radium. Lord Kelvin (William Thomson, 1824–1907) was still arguing for an external source of the energy released by radium and stated that radium was not an element after all, but merely a compound of lead and helium. To prove him wrong, Marie Curie helped by André Debierne undertook the processing of one ton of radiferous residues. After the investment of an

Fig. 5. Marie Curie in 1911.

enormous amount of effort, the team produced a few grams of radium, determined the atomic weight 226.45 (present value 226.0254) and succeeded in preparing metallic radium. The concentration of polonium was also pursued until a fraction containing about 0.1 mg of the element. Spectral analysis revealed a few lines that could be attributed with certainty to polonium 10 years after its discovery. It was the definitive proof for the element and particularly important because it might decay to a stable element. Rutherford commented "it's a matter of very great interest and importance to settle definitively whether polonium changes into lead".

Twelve years after the discoveries of polonium and radium the community of scientists in radioactivity was aware of the importance and urgency of a consensus on an international standard for radium and for units in the field of radioactivity. Rutherford had already questioned Marie who agreed with the proposal. A standard of radioactivity was defined as the amount of emanation in equilibrium with one gram of radium. Marie wished to retain the name "curie" for this unit and was charged with preparing a sample of radium to serve as an international standard of radioactivity. It consisted of 21.99 mg of pure radium chloride (16.75 mg of radium) sealed in a glass vial.

In 1911 Marie Curie was nominee for the Nobel Prize in Chemistry by Gaston Darboux and Svante Arrhenius. On November 10, she was awarded the Prize "in recognition of her services to the advancement of chemistry by the discovery of the elements radium and polonium, by the isolation of radium and the study of the nature and compounds of this remarkable element". December 11, she presented the Nobel lecture "Radium and the new concepts in chemistry". Early in her lecture she stated "Before broaching the subject of this lecture, I should like to recall that the discoveries of radium and polonium were made by Pierre Curie in collaboration with me".

In 1944 the group of Glenn T. Seaborg at the University of California identified the first isotope of the element with atomic number 94, which received the name curium (Cm) to honor the pioneers in radiochemistry.

8. 1921 – Frederick Soddy

In 1921 the Nobel Committee for Chemistry considered that none of the year's nominations met the criteria as outlined in the will of Alfred Nobel. In such a case the Prize can be "reserved" until the following year. The reserved prize for 1921 was awarded in 1922 to Frederick Soddy (1877–1956) "for his contributions to our knowledge of the chemistry of radioactive substances, and his investigations into the origin and nature of isotopes" [30, 31] (Fig. 6).

After graduating from Oxford, Soddy spent a period of 18 months from mid-October 1900 to mid-April 1902 with Rutherford at McGill University in Montreal. The team studied the gaseous emanation of radium and showed that radioactivity involved the disintegration of radioactive atoms to form new elements. Soddy called the process *transmutation*, a term that he borrowed from alchemy. Together Soddy and Rutherford published about ten articles which brought a theoretical explanation of radioactivity and formed the basis of all subsequent investigations. The team also proved the existence of two radioactive series: one starting with uranium and the other with thorium. In March 1903 Soddy returned to England to work with Ramsay [21]. Together they confirmed hat helium was formed during the radioactive decay of radium and evolved in the decay of the radium emanation. In the fall of 1904 Soddy moved to the University of Glasgow, Scotland, where he established the "group displacement law" and proposed the concept of isotopes. Together with John A. Cranston, he was one of the chemists who participated in the discovery of protactinium.

Still in 1904, Soddy lecturing on radium to the Corporation of Royal Engineers, had a premonitory feeling of the dangers of radioactivity and even on the possibility of nuclear weapons: "It is probable that all heavy matter possesses – latent and bound up with the structure of the atom – a similar quantity of energy to that possessed by radium. If it could be tapped and controlled what an agent it would be in changing the world's destiny! 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." [32].

Fig. 6. Frederick Soddy (1877–1956).

In 1911 Soddy published a booklet "The Chemistry of the Radioelements" in which for the first time the chemical inseparability of isotopes was stressed as something fundamentally new and important [33]. From 1904 to 1920 he compiled an Annual Progress Report on radioactivity and atomic theory at the request of the Chemical Society. In 1975 these reports were reproduced and assembled in a volume "Radioactivity and Atomic Theory", an invaluable source of the publications in this period for the historian on radiochemistry and an overall view of the scientific thinking of the time [34].

In 1914 Soddy was appointed Professor of Chemistry at the University of Aberdeen and did not pursue research on radioactivity. In 1919 he was nominated to the chair of Chemistry at Oxford University, a post he held until his retirement in 1937.

It took a long time until Soddy's achievements were recognized by the Nobel Committee. As soon as 1908, Oskar Widman (1852–1930) nominated Soddy together with Rutherford for the Chemistry Prize. Ten years later Soddy was nominated by Rutherford and the German Chemist Wilhelm Schlenk (1879–1943) and again by Rutherford in 1919. In continuation of Soddy's work, the 1922 Nobel Prize in Chemistry was awarded to the British Francis William Aston (1877–1945) for the discovery of isotopes in a large number of non-radioactive elements.

9. 1935 – Frédéric Joliot and Irène Curie

The 1934 Nobel Prize for Physics was not awarded despite 28 nominees among which figured Frédéric Joliot (1900–1958), Irène Curie (1897–1956) and James Chadwick (1891–1974). The following year Chadwick had 13 nominations in Physics and each of the Joliot 7 nominations. The latter had also three nominees in Chemistry including from Ernest Rutherford. Chadwick was awarded the Physics Prize "for his discovery of the neutron" and the Joliot Curie team the Chemistry Prize "in recognition of their synthesis of new radioactive elements". It is amazing that the Nobel Committee confused the term "new radioactive elements" with "new radioactive isotopes of elements".

At the end of the first World War, Irène Curie became her mother's *Préparateur* (personal assistant) and began work on a thesis on polonium [35]. She could not have had a better mentor than Marie Curie who always considered polonium as her own and probably reserved any investigations of the element for her daughter. For the purpose Irène was the first person in France to use a Wilson cloud chamber for recording the range and straggling of particles. She defended her thesis on March 27, 1925 on the radiations emitted by polonium.

Frédéric Joliot who had earned an Engineering Diploma in Physics joined Marie Curie's laboratory in 1925 [36]. He was soon attracted by Irène's exceptional personality and they were married on October 9, 1926. Several years later the surname *Joliot-Curie* came into use, particularly in the media. Numerous associations, schools, streets and official documents, including those of the Nobel Foundation bear

Fig. 7. Frédéric Joliot (1900–1958) and Irène Curie (1897–1956).

the title of Joliot-Curie. Since Pierre and Marie Curie had no sons, adoption of the hyphenated name prevented one of the most famous names in science from disappearing. However, both authors signed their scientific papers, even their joint ones, "Frédéric Joliot" and/or "Irène Curie".

It was not until 1931 that the Joliot couple, married in 1926, began a close collaboration leading within four years to the discovery of artificial radioactivity. Alike the Pierre and Marie Curie team, they were complementary to each other. Although Irène and Frédéric (Fig. 7) were both physicists and chemists, Frédéric is considered as "more" the physicist. With only two exceptions, Irène was the first author in their approximately 40 joint publications.

Frédéric had a particular sense for detecting unexpected phenomena and narrowly missed the discovery of the neutron. In 1933 the Curie couple made a crucial observation: an aluminium foil irradiated with alpha particles from polonium emitted positive electrons – the *positron* – predicted by Paul Dirac (1902–1984; 1933 Physics Nobel laureate) and discovered in 1933 by Carl David Anderson (1905–1991; 1936 Physics Nobel laureate). These findings were extensively discussed in October 1933 at the 7th Solvay Physics Congress attended by a dozen Nobel and future Nobel laureates, including Marie Curie for the last time, and the Joliot couple.

Three months later, Joliot observed that irradiated aluminium was still radioactive several minutes after the polonium source had been removed. He immediately realized the importance of the phenomenon: aluminium was transmuted into a new radioactive substance with decayed with a half life of 2.5 min and emitted positons. The discovery of "A new type of radioactivity" was announced January 15, 1934 at the weekly session of the Académie des Sciences [37]. The Joliot couple had produced and chemically identified ^{30}P, the first artificial radioactive isotope of a stable element and the first positron emitter. The Nobel ceremony was celebrated 18 months after Marie Curies death, 32 years after she shared the Physics Prize with Pierre Curie. It was the second time that Irène participated in a Nobel Ceremony. She was 14 years old when she accompanied her mother 24 years earlier for the awarding of Marie Curie's second Nobel Prize. The couple's brief Nobel lectures were the occasion for each laureate to emphasize the work of the partner although Irène stated "These experiments have been made together by Monsieur Joliot and me, and the way in which we have divided this lecture between us is a matter of pure convenience". Irène, the chemist, was the first speaker on "Artificial production of radioactive elements". She explained the discovery of a new type of radioactivity by emission of positive electrons. Frédéric Joliot, the physicist, followed with "Chemical evidence of the transmutation of elements".

The Joliot couple pursued independent researches, each in a different field and yet were close to further important discoveries. In late 1938, Irène Curie and Paul Savitch (Pavic Savic, 1882–1949) at the *Institut du Radium* in Paris found in neutron irradiated uranium a strongly radioactive substance with a half-life of 3.5 h, carried with actinium. They reported "on the whole its properties are those of lanthanum [element 57] from which, it seems so far, it can be separated only by fractionation". Indeed it *was* lanthanum from the nuclear fission of uranium: the authors were close to the discovery of fission. On his side, Frédéric Joliot at the *Collège de France* proved in 1939 the splitting of uranium by neutrons and demonstrated the development of a divergent nuclear chain reaction two weeks after the outbreak of World War II.

10. 1943 – George de Hevesy

The 1943 Nobel Prize for Chemistry was awarded in 1944 to the Swedish George de Hevesy (1885–1966) (Fig. 8) "for his work on the use of isotopes as tracers in the study of chemical processes". Hevesy had one nomination in Physics in 1942 and a dozen in Chemistry between 1927 and 1944, noteworthy from the Joliot couple in 1936 and from Soddy in 1939. The presentation speech was delivered by A. Westgreen, Chairman of the Nobel Committee for Chemistry in a radio address on December 10, 1943. The situation in Europe at that time hampered travel to Sweden. There was no formal awards ceremony, even though Hevesy was at Stockholm.

In 1911 Hevesy joined Rutherford in Manchester and became interested in radioactive substances. Rutherford had

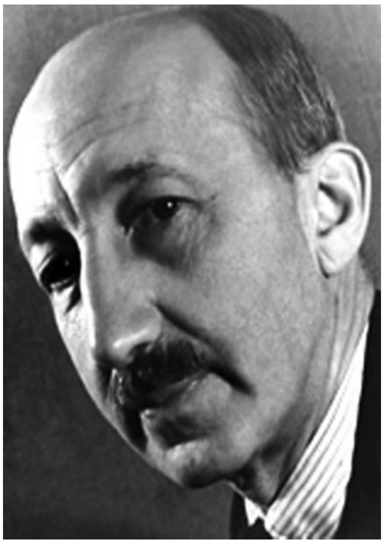

Fig. 8. George de Hevesy (1885–1966).

acquired a considerable amount of "radium D" which was mixed with a large quantity of lead. It was later recognized as a natural radioactive isotope of lead, ^{210}Pb, a daughter product of radium, with a half-life of 22.34 years. At the time isotopes were not yet known. Rutherford asked the young Hevesy "to separate Radium D from all that nuisance lead". Hevesy recalled "Being a young man, I was an optimist and felt sure that I should succeed in my task". One year later he was still at the same point and conceded to have failed but at the same time he had the idea to spike ordinary lead with radium D and to study in this way the properties and behaviour of lead and its compounds.

For the purpose Hevesy moved in 1913 to the Institute of Radium in Vienna where large quantities of radium D were available. Together with the Austrian Fritz Paneth (1887–1958) they discovered the exchange between radioactive and stable lead atoms [38]. Even before the discovery of isotopes by Soddy, the team showed that for all processes the chemical and physical behaviour of these atoms were identical. It was the first application of radioactivity in chemistry and the founding of radioanalytical chemistry [39]. After the discovery of artificial radioactivity it became one of the most common and powerful techniques for the investigation of living and complex systems. Hevesy published over 400 articles and reviews on neutron activation analysis and the use of radioactive isotopes in biology. Besides, he was a chemist with wide interests. In 1923 he discovered with Dirk Coster (1889–1950) the element hafnium (at. number 72), the antepenultimate discovered stable element, while working in Niels Bohr's laboratory in Copenhagen, the former *Hafnia*.

In 1950 Hevesy was awarded the Copley Medal of the Royal Society "for his distinguished work on the chemistry of radioactive elements and especially for his development of the radioactive tracers techniques in the investigation of biological processes". He was particularly proud of the distinction: "The public thinks that the Nobel Prize in Chemistry is the highest honor that a scientist can receive, but it is not so. Forty or fifty received Nobel Chemistry Prizes but only ten foreign members of the Royal Society and only two [Bohr and Hevesy] received a Copley Medal". In 1958 Hevesy received the Atom for Peace Award for the peaceful use of radioactive isotopes.

Hevesy had a chaotic life. Born in Budapest, he studied in Freiburg, where he obtained his doctorate and then worked in Zurich, Karlsruhe, Manchester, Vienna, and Copenhagen. In 1926 he was appointed to the chair of physical chemistry in Freiburg and in 1935 escaped from the Nazis, fleeing first to Denmark and in 1942 to Sweden at the University of Stockholm.

Anecdotes are blowing about this brilliant personality. In his first applied radioactive tracer investigation Hevesy demonstrated that his landlady was recycling food over days. For the purpose he spiked the leftovers of his plate with radium D [40]. When Germany invaded Denmark in World War II, Hevesy dissolved the Nobel Gold Medals of Max von Laue (1879–1960; Physics laureate 1914) and James Franck (1882–1964; Physics laureate 1925) with *aqua regia* to prevent the Nazis from stealing them. After the war he precipitated the gold out of the acid and the Nobel Society recast the Nobel Medal using the original gold.

11. 1945 – Otto Hahn

The 1944 Nobel Prize for Chemistry was announced on November 15, 1945. It was awarded to the German Otto Hahn (1879–1968) (Fig. 9) for the discovery of the fission of heavy nuclei. At that time Hahn had already received 15 nominations in Chemistry since 1914, and 5 nominations in Physics since 1937. Hahn was still detained with other German scientists and could not attend the ceremony. The following year he was remitted the Nobel Prize Medal and the Diploma by King Gustav V of Sweden.

Hahn's interest in radiochemistry began in 1904 while working under Ramsay [41]. In the course of experiments on salts of radium he discovered a substance he called radiothorium (^{228}Th) which at the time was believed to be a new radioactive element. Afterwards he spent several months with Rutherford before returning to Germany where he collaborated with Emil Fischer, the second Chemistry Nobel laureate. Within a few months he discovered further radioactive substances, mesothorium I (^{228}Ra) and II (^{228}Ac), as well as ionium (^{230}Th), the mother substance of radium.

On September 28, 1907 Hahn got acquainted with the Austrian Lise Meitner (1878–1968) (Fig. 10) who become later the first woman full professor in Germany at the University of Berlin [42]. It was the beginning of a thirty-year collaboration and lifelong close friendship between the two scientists. Ten years later the team isolated a substance of long-lived activity which they named *proto-actinium* (now protactinium), the element with atomic number 91 [43].

As soon as 1914, Hahn was nominated to the Chemistry Nobel Prize by the 1905 laureate Adolf von Baeyer. In 1912 he became head of the Radioactivity Department of the newly founded Kaiser Wilhelm Institute for Chemistry in Berlin, since 1956 the "Otto Hahn Building of the Free University".

In 1933 Hahn demonstrated the use of radioactive tracers in chemistry in a book entitled "Applied Radiochemistry" based on lectures given when he was a visiting professor at Cornell University in Ithaca, NY. It was one of the first mentions of "radiochemistry". In 1966, Glenn T. Seaborg recorded (excerpts): "As a young student at the University

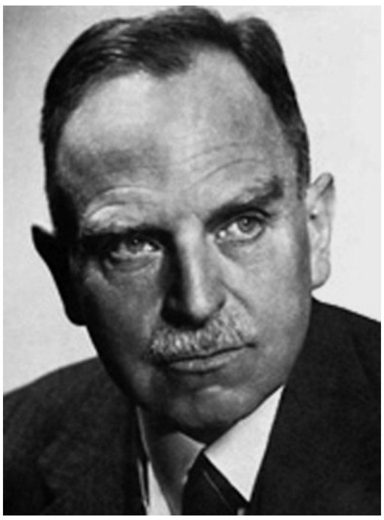

Fig. 9. Otto Hahn (1879–1968).

Fig. 10. Lise Meitner (1878–1968).

of California at Berkeley in the mid 1930s and in connection with our work with plutonium a few years later I used Hahn's book *Applied Radiochemistry* as my bible. It set forth the laws for the co-precipitation of minute quantities when insoluble substances were precipitated from aqueous solutions. I doubt that I have read sections in any other book more carefully or more frequently. In fact, I read the entire volume repeatedly and I recall that my chief disappointment with it was its length. It was too short".

In 1934 Hahn and Meitner turned to the search of transuranium elements initiated by Enrico Fermi (1901–1954) when it was believed that these elements could be formed in uranium irradiated with neutrons. They found a great number of radioactive atoms including isotopes of barium. The crucial experiment was done in Berlin in December 1938 by Otto Hahn and Fritz Strassmann (1902–1980) while Lise Meitner had escaped to Stockholm. It was entitled "On the detection and characteristics of the alkaline earth metals formed by irradiation of uranium with neutrons" in *Naturwissenschaften* early 1939. Meitner and her nephew Otto Robert Frisch (1904–1979) interpreted the results as "Disintegration of uranium by neutrons: a new type of nuclear reaction" published February 11, 1939 in *Nature* [44]. It was confirmed experimentally by Frisch a few weeks later as "Physical evidence for the division of heavy nuclei under neutron bombardment" [45]. The uranium nuclei had split to form barium and krypton accompanied by the ejection of several neutrons and large amounts of energy; Frisch coined the word "fission". News of the splitting of the atom was brought by Niels Bohr (1885–1962) to scientists in the United States and ultimately resulted in the Manhattan Project.

Hahn received many governmental and academic awards. He was elected member or honorary member in 45 academies and scientific institutions. In 1966 he received with Lise Meitner and Fritz Strassmann the Enrico Fermi Prize, the first time the Prize had been awarded to non-Americans.

Hahn's name was ephemeral in the chart of elements. In 1970 the American Chemical Society proposed hahnium (Ha) for element 105 which is now dubnium, Db. In 1997, hahnium was again recommended for element 108 with symbol Hn, but eventually in 1992 the element became officially hassium, (Hs), derived from the Latin name where the institute is located (L. *hassia*, German *Hessen*).

Lise Meitner was overlooked by the Swedish Academy despite 9 nominations in Physics between 1937 and 1949 and 14 nominations in Chemistry between 1924 and 1948. To say the least, she was cited in the presentation speech of Otto Hahn for the 1944 Nobel Prize. A. Westgren, Chairman of the Nobel Committee for Chemistry said "In collaboration with Lise Meitner, with whom he had worked for nearly thirty years, Hahn studied from 1936 to 1938 the products obtained by projecting neutrons onto the heaviest elements, thorium and uranium. According to Fermi, elements would appear which would form a continuation of the Periodic Table of the elements. Hahn and Meitner believed they could confirm".

In private, Einstein would some times express the opinion that Lise Meitner was "the German Marie Curie" and "more talented physicist than Marie Curie herself " The two women and the Joliot couple met once in 1933 at the seventh Solvay Conference when Meitner had already received 5 nominations to the Chemistry Nobel Prize.

12. 1951 – Edween M. McMillan and Glenn T. Seaborg

The 1951 Nobel Prize for Chemistry was awarded to Edwin M. McMillan (1907–1991) (Fig. 11) and Glenn T. Seaborg (1912–1999) (Fig. 12) for their discoveries in the chemistry of the transuranium elements [46].

McMillan was an "accelerator physicist" alike the 1951 Physics laureates John D. Cockroft (1897–1967) and Ernest T. S. Walton (1903–1995). He joined the group of Ernest Lawrence at the University of California and the Berkeley Radiation Laboratory when it was founded in 1934. He discovered the isotopes ^{15}O with Stanley Livingston and ^{10}Be with Samuel Ruben. In 1940 McMillan and Philip Abelson (1913–2004) synthesized the first isotope of neptunium (at. number 93), the first transuranium element, while conducting a fission experiment on uranium 238. Later McMillan had the idea of "phase stability" which led to the invention of the synchrotron and the synchro-cyclotron for which he could be eligible to a Nobel Prize in Physics.

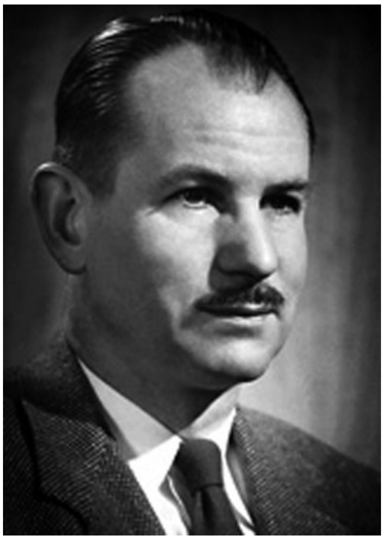

Fig. 11. Edwin M. McMillan (1907–1991).

Fig. 12. Glenn T. Seaborg (1912–1999).

Fig. 13. Willard F. Libby (1908–1980).

Seaborg's main contribution to chemistry is the concept of actinide elements, the 15 elements atomic numbers from 89 to 103 corresponding to the filling of the $5f$ electron shell. Lawrencium (at. number 103), a *d*-block element, is also generally considered an actinide. Seaborg remembered "In 1944, I got the idea that may be these elements were misplaced in the Periodic Table. Perhaps the new heavy rare earth series should start back at thorium with actinium as its prototype, thus dubbing the collection of the actinide series. When we tried this idea, we found that it was right" [46].

Seaborg was the principal or co-discoverer of nine elements: plutonium, americium, curium, berkelium, californium, einsteinium, fermium, mendelevium, and nobelium and predicted chemical properties and electronic structure of the transactinide elements and the nuclear stability of superheavy elements beyond. The element 106 discovered in 1974 was publicly named seaborgium, a unique denomination of a living scientist. Incidentally, einsteinium (at. number 99) and fermium (at. number 100) were also named while the eponymous discoverers were living but the names were kept under the Cold War.

After sharing the Nobel Prize with McMillan, Seaborg received about 50 honorary doctorates and numerous awards and honors. He was listed in the Guinness Book of World Records as the person with the longest entry in *Who's Who* in America.

13. Nobel Prizes in Applied Radiochemistry

Arthur C. Wahl and Norman A. Bonner in the Preface of their book "Radioactivity Applied to Chemistry" published in 1951 commented "During and after World War II an increasingly large number of chemists were becoming interested on the applications of radioactivity" [47]. They could not foresee that radiochemistry would lead to several Nobel Prizes in the following years.

In 1960 the Chemistry Prize was awarded to Willard F. Libby (1908–1980) (Fig. 13) "for his method to use carbon-14 for age determination in archaeology, geology, geophysics and other branches of science" [48]. Regrettably, Martin D. Kamen (1913–2002) and Sam Ruben (1900–1943), discoverers of ^{14}C in 1940, were not nominated nor even mentioned in Libby's Nobel conference [49, 50]. Libby was the founder of "hot atom chemistry", the investigation of reactions of energetic atoms produced in nuclear reactions and radioactive decays first reported by Leo Szilard and T. A. Chalmers in 1934 [51].

In 1961 the Chemistry and Physics Prizes relied essentially on researches with radionuclides. The Chemistry Prize was awarded to the American Melvin Ellis Calvin (1911–1997) (Fig. 14) for his research on the carbon dioxide assimilation in plants performed with carbon 14. The German Rudolf Ludwig Mössbauer (1929–2011) (Fig. 15) received the Physics Prize for his researches on the resonance absorption of gamma radiation and the discovery in this connection of the effect which bears his name. Various nuclides, particularly ^{57}Co, led to the development of Mössbauer spectroscopy, a powerful method for the investigation of chemical bonds of iron, tin and other elements. Radiochemists developed "Mössbauer emission spectroscopy" for the investigation of the chemical effects of radioactive de-

Fig. 14. Melvin E. Calvin (1911–1997).

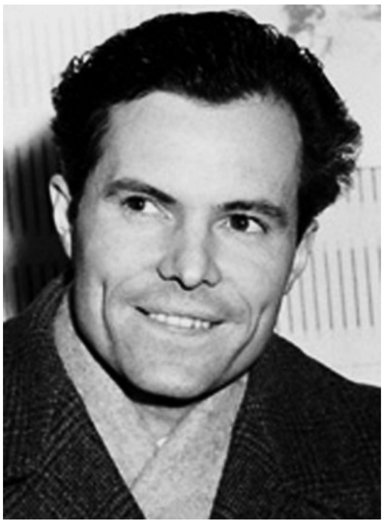

Fig. 15. Rudolf Mössbauer (1929–2011).

Fig. 16. Rosalyn Yalow (1921–2011).

cays. Rosalyn Yalow (1921–2011) (Fig. 16), medical physicist, was awarded the 1977 Prize in Physiology or Medicine for development of the radioimmunoassay (RIA) technique, a radioisotope tracing technique that allows the measurement of tiny quantities of various biological substances in human blood as well as a multitude of other aqueous fluids [52].

F. Sherwood Rowland (b. 1927), (Fig. 17) shared the 1995 Chemistry Prize with Paul J. Crutzen (b. 1933) and Mario J. Molina (b. 1943) "for their joint work in atmospheric chemistry, particularly concerning the formation and decomposition of ozone". Rowland discovered that manmade organic compounds, such as chlorofluorocarbons (CFC) released in the stratosphere are decomposed by solar radiation. They give rise to highly reactive Cl and chlorine monoxide (ClO) species which destroy ozone by catalytic reactions. The decrease in the Earth's ozone layer has far-reaching environmental consequences, notably an increase in skin cancers [53]. Rowland was already a famous radiochemist for his work on the chemistry of "hot" atoms undertaken in the 1950's with Willard F. Libby. In 1991 he was listed as one of the "1000 Makers of the Twentieth Century" by the London *Sunday Times* [54].

Fig. 17. F. Sherwood Rowland (b. 1927).

14. Nobel nominees, but not winners

The initial work of the Curies provoked immediate researches on radioactivity outside of France. In Germany Julius Elster and Hans Geitel published about 30 articles in the field and received 7 nominations for the Physics Prize between 1904 and 1911 [13]. Pierre Curie commented in 1898 "Elster and Geitel are certainly persons who most thoroughly investigated the questions of uranic rays and I would not be surprised if they soon produced excellent results".

André Debierne, discoverer of actinium and collaborator of Marie Curie with whom he determined the atomic weight of radium and the first spectrum of polonium, was nominated in 1925 by the French chemist Albin Haller (1849–1925), recipient of the Davy Medal in 1917 and President of the *Académie des Sciences* in 1923.

William Crookes (1832–1919) [55, 56], discoverer of thallium in 1861, missed the Nobel award despite nine nominations to the Physics or Chemistry Prize, but was awarded the Copley Medal of the Royal Society of London "for his long-continued researches in spectroscopy, on electrical and mechanical phenomena in highly-rarefied gases, on radio-active phenomena and other subjects". Crookes was the leading British spectroscopist of his day, participating in the discovery of argon and helium. About 1875 he invented the "Crooke's tube" later used in experiments leading to the discovery of X-rays by W. C. Roentgen in 1895 and the electron by J. J. Thomson in 1897.

At the age of 67, Crookes became interested in radioactivity. He observed that when radiations ejected from radioactive substances impinge upon zinc sulfide, each impact is accompanied by a minute scintillation, an observation which is the basis of one of the most useful detectors in radioactivity. On the course of the treatment of pitchblende with the intention to extract polonium and radium he found to his astonishment that the uranium did no longer blacken the photographic plate. Retracing his steps, he discovered the fugitive activity in the aqueous portion of the ether extraction and concentrated it in a small quantity of intensely radioactive precipitate. He concluded "the supposed radioactivity of uranium and its salts is not an inherent property of the element, but is due to the presence of a foreign body" which he called uranium X, the unknown substance in uranium, now ^{234}Th (half-life 24 days) [57]. This discovery was considered by the Nobel Committee as Crookes' most relevant one. If Crookes had tested his purified uranium a month or two later, he would have found that it was again intensely radioactive, while the uranium X had lost nearly all of its capacity to blacken a photographic plate. Crookes narrowly missed the discovery of atomic transmutation as he did also for radioactivity: he once sent back to a manufactor a batch of photographic plates stored close to uranium, complaining that they were fogged.

Kasimir Fajans (1887–1975), of Polish origin, was nominee in 1928. He formulated the radioactive displacement law with Frederick Soddy. In 1913 together with Oswald Helmut Göhring he discovered the radionuclide of a new element which was later named protactinium. He formulated "Fajans' rules" of inorganic chemistry which are used to predict whether a chemical bond will be covalent or ionic.

The Noddack couple, Ida née Tacke (1896–1978) and Walter (1893–1960), received several nominations between 1932 and 1937. In 1925 the Noddack discovered rhenium, atomic number 75, and announced the discovery of the element with atomic number 43 (technetium) which they named masurium. The lasting claim of this erroneous discovery tarnished the couple's fame. On the other hand, Ida Noddack criticized correctly Fermi's announcement in 1934 of the production of transuranic elements in the neutron bombardment of uranium: "It is conceivable that the nucleus breaks up into several large fragments, which would of course be isotopes of known elements but would not be neighbours of the irradiated element" [57]. However she did not offer any explanation to sustain the proposal.

The Austrian Friedrich Adolf Paneth (1887–1958) received five nominations between 1927 and 1939 including one from Soddy and further six proposals in 1948. He was a prolific forerunner in the chemistry of minute amounts of radioelements, the applications of radioactive indicators and founder of cosmochemistry. Besides, he was a scholar with a prodigious culture, with dissertations, among others, on "Goethe's scientific background" and "Science and Miracles" [31].

15. Forgotten discoverers of radioelements

Discoveries of several radioactive elements were not submitted to Nobel Committees: technetium (at. number 43, Carlo Perrier and Emilio Segré in 1937); francium (at. number 87, Marguerite Perey in 1939); astatine (at. number 85, Dale R. Corson, Kenneth R. Mackenzie and Emilio Segré in 1940); promethium (at. number 61, Jacob A. Marinsky, Lawrence E. Glendenin and Charles D. Coryell in 1945).

16. Conclusion

The 1951 Nobel Prize in Chemistry awarded for the discoveries of the first two transuranium elements is presently the last one awarded in the field of fundamental radiochemistry. The expansion of the Periodic chart of the elements was the occasion to pursue the recognition of Nobel pioneers in radiochemistry with curium, atomic number 96, in 1944; rutherfordium, atomic number 104, in 1964 and seaborgium, atomic number 106, in 1974. Last but not least, the forgotten Lise Meitner joined in 1982 the "immortal Nobel laureates" with meitnerium (Mt) the element with atomic number 109 first synthesized in 1982. The latest IUPAC chart of elements ends with copernicium (Cn) (at. number 112) while syntheses of elements 113 up to 118 were reported. The present status of researches on transuranium elements was reported in a special issue of Radiochimica Acta, "Heavy elements" (2011) [58].

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