Chapter 6 Radioactivity

...und nun ging der Teufel los.. (Wilhelm Röntgen)

6.1 The Revival of Inorganic Chemistry

At the end of the nineteenth century, inorganic chemistry had lost most of its importance as ancestor of all other disciplines in which chemistry was evolving and was at the point of becoming more of an introductory teaching vehicle than a busy line of research as in the past. The study of the individual elements started to appear to the new generations of chemists as poor relative to the two great branches of chemistry, organic and physical chemistry. These instead were witnessing a period of great splendor and intense development, the first thanks to the formidable successes of organic synthesis and the second for the rigorous theoretical organization in which it had framed the structure of the molecules and the identification of their physical properties.

The work of the many "element hunters," who in the previous century had achieved fame and celebrity by digging in the northern regions of Sweden and Germany for strange ores from which new exotic elements were extracted through a patient and boring series of analytical procedures, seemed already bypassed by a growing interest in the interaction between electromagnetic fields and matter. This new field had found its peak of fame in the discovery of x-rays and of the fluorescence that this new source of radiation was able to induce in many materials. It was only thanks to the perseverance of a young Polish girl, Manya Sklodowska, who arrived in Paris to study physics at the Sorbonne, that atomic nuclei started to whisper again in their faint voice in an old laboratory of Rue Cuvier in the Latin Quarter of Paris; this was thanks to another kind of radiation, this time spontaneously emitted by some uranium compounds. From there this voice became a stronger and stronger roar, being heard throughout the world and giving rise to the new fascinating field of nuclear chemistry, later leading to the discovery of atomic energy and to its use for both good and evil.

6.2 The Curie Couple

The discovery of radioactivity started from the observation made in 1896 by Antoine Henri Becquerel that potassium uranyl sulfate $K_2UO_2(SO_4)$ spontaneously emitted a strange radiation capable of affecting a photographic plate. Becquerel came from a family of physicists; the grandfather, Antoine César (1788–1878), developed a method to extract metals from their minerals and the father, Alexander Edmond (1820–1891), studied solar radiation, the fluorescence and phosphorescence of minerals, and was a leading authority in Europe on the phosphorescence of solids. Just from his father, Antoine inherited a rich collection of fluorescent and phosphorescent minerals that later turned out to be precious for his research activities.

Antoine Henri Becquerel (1852–1908) obtained the title of engineer at the Ecole Nationale des Ponts et Chaussées in 1877 and a Ph.D. in physics in 1888. In a short while, he became professor of applied physics at the Museum of Natural History in 1892, at the Ecole Polytechnique in 1895, and later at the University of Paris. In a session of the Académie des Sciences, of which he was a member, he had the chance to listen to an enthusiastic lecture from the mathematician Henri Poincare´ (1854–1912) on the discovery of x-rays, which occurred almost by accident by observing the fluorescence that they produced on striking the walls. Poincaré had received a letter from Wilhelm Conrad Röntgen, a scientist he had never met, with some astonishing photographs of bones made using the newly discovered radiation. In his lecture, Poincaré advanced the hypothesis that x-rays could somehow be related to the phenomenon of phosphorescence and Becquerel, impressed by this idea, decided to restart the studies of his father on induced fluorescence and phosphorescence, using solar light as the exciting radiation.

After having studied several compounds, he realized that fluorescence and phosphorescence were similar physical processes, both stimulated by external radiation. The main difference that he noticed between the two processes was that while fluorescence ceased if the exciting radiation was stopped, phosphorescence continued for a long time.

On 25 January 1896, Antoine Becquerel started the study of induced fluorescence in crystals and on 24 February reported to the Academy that several materials, in particular the crystals of potassium uranyl sulfate $K_2UO_2(SO_4)_2$, a compound known for its phosphorescence properties, emitted rays that penetrated thick sheets of black paper. Becquerel's approach to the study of the induced phosphorescence was to expose the sample to solar light and position it on a photographic plate wrapped in light-tight black paper, leaving the experiment on a windowsill where, according to his hypothesis, the sunlight would stimulate the mineral to glow, emitting the unknown x-rays. Wednesday, 26 February, and Thursday, 27 February, were days without sunshine in Paris and Becquerel, being unable to use solar energy for his experiment, decided to position the sample on a package of photographic plates wrapped in black paper in a drawer in his laboratory and left it there. On 1 March he developed the plates and found that these had been impressed by an invisible radiation spontaneously emitted by the crystals without exposition to solar light. It is even possible, as told in some records of Becquerel's experiment, that he simply forgot the sample positioned in the drawer. In any case, when he developed the plate and saw the shadow of the crystals impressed on it, he reached the wrong conclusion that the solar radiation, adsorbed by the crystals, was transformed in x-rays.

The first of March was actually a Sunday and it is hard to believe that he went back to his laboratory without a serious reason to carry out an operation of little interest that his colleagues of the Academy had practically ignored and that did not seem important compared to Röntgen's discovery. In reality, Becquerel knew that one of the pioneers of photography in France, Claude-Félix-Abel Niepce de Saint Victor (1805–1870), had already observed in 1867 that uranium salts weakly blackened photographic plates even when wrapped in several sheets of black paper. The observations of Niepce were reported in a book written by Becquerel's father that he undoubtedly read. It is therefore highly possible that, remembering Niepce's findings, he went back to the laboratory to examine the plates and to make sure that the phenomenon described by Niepce was real.

After a few days, Becquerel reported his discovery to the Académie des Sciences (Becquerel [1896a](#page-28-0), [b](#page-28-0)), but the news was almost completely neglected by the members of the Academy, excited by the discovery of x-rays to which Becquerel's rays did not seem comparable. In addition, after Röntgen's discovery, the hunt for all kind of invisible rays emitted by matter had started in the scientific community, leading to a plethora of phenomena, often purely imaginary, that gave rise to quite a bit of confusion and overall to much diffidence.

Among the several inventors and professed physicists impressed by Röntgen's discovery, the French Gustave Le Bon (1841–1931) succeeded more than the others in attracting the attention of the press and even the sympathy and benevolence of important scientists such as Poincaré. Le Bon, social psychologist, sociologist, and amateur physicist, reported in 1896 having discovered a "black light," a new type of invisible electromagnetic radiation different, but somehow correlated to, x-rays. Even if he did not receive too much attention in the physics community, his ideas on matter, radiations, and the ether stirred the interest of a wide audience starting to be aware of the imminent revolution against positivism. His book $L'E$ volution de la matière published in Paris by Flammarion in 1905, in which he maintained that the matter is unstable and transforms slowly but inexorably into a radiation, had as many as 12 editions in France.

Less lucky was the physicist René Blondlot (1849–1930), professor at Nancy, known for his research on electromagnetism, on Maxwell's theory, and for having realized in 1891 the first measurement of the propagation speed of radio waves. Blondlot added to Le Bon's fantasies a famous mistake of experimental physics, announcing to the Académie des Sciences in 1903 the discovery of a new type of rays that he named N rays in honor of Nancy. In a very short time, several confirmations of the existence of the N rays appeared in the scientific literature and the Académie even awarded him a 20,000 francs prize. Unfortunately for him in 1904 the American physicist Robert William Wood (1868–1955), professor of optics at the Johns Hopkins University, visiting Blondlot's laboratory at the behest of the journal Nature, revealed (Wood [1904\)](#page-31-0) that Blondlot's discovery was a phenomenon of collective self-deception and imagination and not a physical process since he had verified that the N rays were detected by Blondlot's coworkers even when he had surreptitiously removed a prism from the optical path! Despite the indifference and disinterest of the members of the Académie, Becquerel continued to work for some time on the radiation emitted by uranium salts, proving with the aid of a magnetic field that it consisted of charged particles. Later he devoted himself to the study of the effect of magnetic fields on spectral lines, an effect discovered in 1896 by Pieter Zeeman (1865–1943) at Leyden, and presented a series of communications on the Zeeman and Faraday effects to the Académie. Even his son, Jean-Antoine-Edmond Becquerel (1878–1953) followed in the footsteps of his father although with less success.

If the Academicians did not play too much attention to Becquerel's discovery, his work was continued by a young Polish woman, Marie Sklodowska, and by her husband, Pierre Curie. With their research they created a new branch of chemistry, radiochemistry, procuring for Becquerel the honor of the Nobel Prize in physics in 1903.

Pierre Curie (1859–1906), who had the position of instructor at the Municipal School of Industrial Chemistry and Physics in Paris, was an already established physicist when he met his future wife. In 1880, together with his brother Jacques, he discovered piezoelectricity (Curie and Curie [1880](#page-28-0)) and was internationally known for having found that the magnetic susceptibility of paramagnetic materials was inversely proportional to the absolute temperature (Curie–Weiss law) as well as for having identified the temperature, the Curie point, above which the magnetic properties of a material disappear (Curie [1895\)](#page-28-0). Despite these significant scientific results, he had not received the respect from his peers to match the level of his research, mostly because of his reserved and shy nature, alien to academic politics, but also because he had not yet written his Ph.D. thesis that he completed in March 1895, a few months before getting married.

Marya (afterward changed to Marie) Sklodowska (1867–1934) was born in Warsaw, Poland, at that time part of the Russian empire. In 1891, she followed her elder sister Bronisława to go to study in Paris, where she graduated at the Sorbonne in physics in 1893 and in mathematics in 1894. To support herself at the University she started, under the direction of the physicist Gabriel Lippmann (1845–1921), to study the magnetic properties of steel, thanks to a fellowship financed by the society for the support to the national industry.

Soon after their marriage in July 1895, the Curie husband and wife started to work together on the magnetism of metals in the small and badly equipped laboratory that Pierre had directed since 1882. In 1897, Marie completed her work on steel magnetism and started to look for a new field for her Ph.D. thesis. The discovery of the strange radiations emitted by uranium salts, made 1 year before by Becquerel, fascinated Marie who decided to select this completely new, virgin, and very promising topic for her Ph.D. thesis. Using an electrometer based on the piezoelectric effect, invented by Pierre and by his brother Paul-Jacques (1856–1941), Marie dedicated herself to the determination of the conductibility of air exposed to uranium salt radiation, a technique that Becquerel had already used with success to evaluate radiation intensity. The apparatus employed by Marie consisted essentially of a plate condenser, on which the finely powdered active material was spread, making the air between the plates a conductor of electricity. In order to measure the conductivity, the plate was raised to a high potential by connecting it to the pole of a battery of small accumulators. Marie found that the emission of rays was proportional to the uranium content of the material, constant over time, and independent of temperature and the illumination of the sample. She therefore started to test all available compounds to understand which of them emitted the unknown rays.

In this research, she was helped by several chemistry colleagues who supplied samples of rare earths, and in April 1898 she discovered that the same radiation was also emitted by thorium compounds, an element identified by Berzelius in 1828. By systematically investigating different compounds, she soon reached the conclusion that the intensity of the emitted radiation was independent of the chemical composition and depended only on the quantity of uranium or thorium present in the samples and understood that the capability of the emitted rays was a property specific to the atoms of uranium and thorium. The immutable atoms of the Greek philosophers had finally started to speak and to reveal their complex internal structure.

On 17 February 1898, the Curie couple examined samples of some uranium ores: pitchblende, a black pitch like mineral from the Joachimsthal mines in Bohemia, and chalcolite (also known as torbernite). Both minerals contained uranium, thorium, and rare earth oxides, and produced an ionization of the air greater than that of uranium oxide. Pitchblende was four times as active as uranium itself, and chalcolite twice as active.

After repeated measurements, yielding always the same result, Marie Skłodowska-Curie, as she liked to be called to underline her Polish origins, reached the conclusion that unknown compounds, more radioactive than uranium, were present in the ores. Pierre, once he realized that what she observed was not a spurious effect and that his wife had in her hands an important discovery, abandoned his own research on crystal symmetry and magnetism to dedicate himself to identifying, with her, these new unknown compounds.

Marie Curie collected in her memoirs a detailed description of this famous period of her life, of the squalid laboratory with the floor in a precarious condition, of the poor available equipment, of the cold that they suffered in winter and the asphyxiating heat in summer, and, above all, of the back-breaking work of purification and extraction of the radioactive fractions from tons of raw material. Her daughter Eve Curie (1904–2007) reconstructed with passion the fascinating atmosphere of this romanced and romantic period of her mother's life in a well-known book (Curie [1938\)](#page-28-0).

In this period, the Curie couple changed to analytical chemists and, using separation techniques involving an infinite series of interminable sequences of fractional crystallizations, succeeded in isolating from the pitchblende ores several

fractions, one of which, rich in bismuth, and another, rich in barium, were both strongly "radioactive."

From the bismuth-rich fraction they obtained, at the end of June 1898, a new substance, chemically similar to bismuth, and about 400 times more radioactive than uranium. On 18 July 1898, they published in Comptes Rendus a paper entitled Sur une substance nouvelle radioactive, contenue dans la pechblende (Curie and Curie [1898](#page-28-0)) in which they suggested for the new element the name of polonium, in honor of Marie's native country. In this paper, the term *radioactive* was mentioned for the first time, a term coined by Marie to indicate the emission of the unknown rays that Becquerel had named "U rays."

The true nature of these U rays was discovered in 1898 by Ernest Rutherford who showed that they consisted of two kinds of particles that, with a minimum effort of imagination, were called α and β rays. In the same period of time, the U rays were studied in detail by Hans Mayer [\(1904](#page-30-0)) as well as by Stefan Meyer and Egon von Schweidler ([1905\)](#page-30-0).

After few months, the Curies identified in the barium rich fraction, in collaboration with the chemist Gustave Bemont (1857–1937), coworker of Pierre, the existence of a second radioactive element for which they suggested the name radium. The discovery of radium was published on 26 December with the title Sur une nouvelle substance fortement radioactive, contenue dans la pechblende (Curie et al. [1898\)](#page-28-0).

At the beginning of 1900, Pierre brought to his research a young chemist André Debierne (1874–1949), assistant at the laboratory of Charles Friedel of the Ecole de Chimie et Physique. Debierne was a short, stocky, and bald introvert personality who had relatively little credit in the international physics community. Nevertheless, he became an important friend of the Curie family and intensively collaborated with them in their research on unknown radioactive elements.

Starting from 1900, Marie obtained a chair at the École Normale Supérieure of Sevres. In 1902, she was able to announce success in isolating 0.1 g of pure radium chloride from more than a ton of pitchblende, obtained thanks to the courtesy of the geologist Eduard Suess (1831–1914) born in London but Austrian naturalized and a member of the Austrian Academy of Sciences. In 1902, in collaboration with Debierne, Marie isolated pure radium by electrolysis of the chloride using a mercury cathode and distilling off the mercury in a hydrogen atmosphere. From a measurement of the molecular weight and from the emission spectrum, it was easy to prove that it was really a new element. An important contribution to the preparation of appreciable amounts of pure radium came from the work of another Austrian scientist, the physicist Stefan Meyer (1872–1949), director of the Institute for Radium Research in Vienna. Meyer had started his scientific activity as assistant to Boltzmann at the Institute for Theoretical Physics, University of Vienna, working on the magnetic permeability of liquids. Influenced, however, by a lecture of Friedrich Oskar Giesel, a pioneer in the research and production of radium, he decided to work on radioactivity and, in collaboration with Egon von Schweidler $(1873-1948)$, was able to show that the β rays could be deflected by magnetic fields. At the beginning of the twentieth century (1910), the Institute for Radium Research in Vienna was opened thanks to funds donated in 1908 by the Austrian industrialist Karl Kupelwieser, and Meyer became its first acting director. By request of the Austrian Academy of Sciences, Meyer organized, with the assistance of technicians of the Auer von Welsbach chemical plant, the production of 4 g of radium by extraction from pitchblende obtained from the Sankt Joachimsthal mines, located in Austria-Hungary. The radium – which Meyer shared with the Curies in Paris, Rutherford in Manchester, and Ramsey in London – made him a key figure in the research on radium.

The fact that materials containing radium spontaneously emitted light, presented by the Curies at the First International Physics Conference held in Paris in 1900, excited the imagination of the public, making radioactivity the most popular branch of science of the time and giving rise to a series of commercial activities and of mass media interest, intimately connected to an increase of its public visibility.

In June 1903, Marie Skłodowska-Curie obtained a Ph.D. in physics and in the same year she and her husband Pierre Curie shared with Becquerel the Nobel Prize in physics. One half of the Prize was assigned to Becquerel for the discovery of radioactivity and the other half to Pierre and Marie for the discovery of polonium and radium. With the Nobel Prize, Marie and Pierre Curie suddenly became famous. The Sorbonne gave Pierre a professorship and permitted him to establish his own laboratory, in which Marie became the director of research.

In January 1904, the industrialist Armet de Lisle, who supported the Curies in the production of radium, opened new industrial installations for producing radium for medical applications. Thanks to De Lisle's help, the Curies were able to obtain larger samples of radioactive material than they would never be able to prepare on their own. In 1904, the journal Le Radium was founded, the first scientific journal devoted to radioactivity, edited by Jacques Danne, an assistant to Pierre Curie, and directed by Henri Farjas. Le Radium initially was intended to be an "instrument" of popularization and research," but after only 6 months it became a strictly academic journal. In the same period, books for a broad public of nonexperts were published by Marie Curie, Untersuchungen über die Radioactiven Substanzen (Braunschweig, Vieweg, 1904), by Henri Becquerel, On the radio-activity of matter (from The Smithsonian Report for 1902, Washington, 1903), by Foveau de Courmelles, Les applications médicales du radium (Imprimerie P. Orsoni, Paris, 1904), and by Alphonse Berget, Le radium et les nouvelles radiations: que faut-il en penser? Que faut-il en attendre? (Paris, G. Charpentier 1904).

On 19 April 1906, a tragedy struck the Curie family. Pierre was killed in a street accident, struck by a horse-drawn car while walking in heavy rain across Rue Dauphine near the Pont Neuf. Marie, devastated by the death of her husband, was left alone with two daughters, Irène 9 and Eve 2 years old. Fortunately, the physics department decided to entrust her with the chair created for Pierre Curie and also to appoint her as director of the laboratory.

Marie was the first woman to obtain a Ph.D. in science in 1908, the first woman to become professor of physics at the Sorbonne, the first woman invited to a Solvay conference, and the first scientist to obtain two Nobel Prizes, the second for chemistry being awarded to her in 1911. In spite of these important recognitions and of the great international success of her researches, Marie had to face at the end of 1910 and for the whole 1911 a ferocious denigrating campaign in the French press. At the beginning of the nineteenth century, the nationalist and chauvinist French society, fueled by gloomy currents of xenophobia, anti-Semitism, and antiscientism, would not easily accept that a Polish woman, with a last name like Sklodowska and an additional first name "Salomea," suggesting a Jewish origin, furthermore connected to a group of intellectual supporters of the cause of the traitor Dreyfus and suspected of antinationalistic political positions, could be considered a true representative of the great French culture. This disparaging campaign, directed by the journalist Léon Daudet in the journal L'Action Française, became more intense when a sentimental relationship of about a year's duration between Marie and the physicist Paul Langevin (1872–1946), a former student of Pierre Curie, was made public, portraying Marie as a homewrecker. This resulted in a great press scandal that excited the "prudery" of affluent French society.

This "Langevin affair" rapidly became a national scandal that culminated in a pistol duel, fortunately without consequence, between Paul Langevin and Gustave Téry, editor of the journal L'Oeuvre who had published some extracts of compromising letters of Marie to Langevin, stolen from the Langevin house. The matter deteriorated further when Téry accused Langevin of being a cad and a scoundrel so that, to defend his honor, Langevin was forced to challenge him to a duel. The duel with pistols at a distance of 25 m was organized for the morning of 26 November 1911. However, it turned into a real farce. The inexperienced Langevin even had a difficult time finding seconds for the day of the duel, since his colleagues were afraid of being involved in this unpleasant story and only at the last moment did he succeed in convincing two friends, in particular the mathematician Paul Painlevé, who later would become prime minister of the French Republic, to assume this embarrassing job. The morning of the duel was gray and foggy. Langevin arrived first and Téry with his entourage arrived after a short while. Painlevé who had been chosen by lot to direct the procedure but had no practice of duels, after having asked the combatants if they were ready, to the general surprise, without waiting, counted with a loud voice one, two, three, fire, before the two adversaries could be ready. Langevin who was just as incompetent raised his arm with the pistol, but, seeing that Téry was not yet ready, lowered his arm again. At this point, Téry said that he had no intention of depriving France of one of her best physicists and he too lowered his arm. Painlevé then quickly decided that the duel was over and in a general silence the two combatants went back home.

However, the situation became more and more embroiled when, due to the discomfort created by a scandal that had assumed international dimensions, Svante Arrhenius, member of the Nobel committee, sent a letter to Marie suggesting that she should refuse to go to Stockholm to receive the prize until the situation could be fully settled. Marie obviously answered rather truculently that she had received the Nobel Prize for the discovery of plutonium and radium and that the evaluation of the Nobel Academy could not be influenced by gossip relating to her private life.

When Albert Einstein heard the story of the Langevin affair, he wrote to Marie:

I will always be grateful that we have people like you and Langevin among us.... If the rabble continues to be occupied with you, simply stop reading that drivel. Leave it to the vipers it was fabricated for.

On 11 December, Marie, holding her head highly, pronounced the Nobel speech at Stockholm, and after the ceremony dined at the Royal Palace with the King of Sweden, Gustaf V. However, the stress provoked by this unhappy series of events gave rise to a strong nervous breakdown for which she was after hospitalized and for almost a year unable to go back to work.

For the rest of her life Marie dedicated herself to the development of the use of radium for the treatment of cancer, an important medical application that Pierre had predicted once he had realized that the radiations emitted by radioactive compounds could destroy organic molecules and in particular could be used to destroy malignant tumors (Quinn [1995\)](#page-30-0).

6.3 Hunting for New Radio Elements

With her research Marie Curie opened a new chapter of inorganic chemistry that was soon found to be extremely rich for important developments, giving rise, soon after the discovery of radioactivity, to a large amount of new research that flourished in few years all over the world.

In France, André Debierne, who had dedicated himself to the research of an element that he suspected should have been present in an iron rich-fraction extracted from pitchblende, discovered in 1899 a new radioactive element, actinium, and presented his discovery in three papers (Debierne [1899,](#page-29-0) [1903](#page-29-0), [1904\)](#page-29-0). At Marie's death in 1934, Debierne became director of the Curie laboratory despite the fact that he did not have the international renown expected for the successor of a personality like Marie Curie.

In 1898, in Germany Gerhard Carl Schmidt (1864–1949), professor at the University of Erlangen, discovered slightly before Marie Curie the radioactivity of thorium. Schmidt reported his results to the Deutsche Physikalische Gesellschaft in Berlin on 4 February 1898 and published a paper on this subject on 24 March of the same year (Schmidt [1898](#page-31-0)), a few months before the communication presented by Marie Curie on 12 April at the Académie des Sciences of Paris. Unfortunately for him, Marie Curie's name had already reached such a high level of popularity in the physics community that it obscured his priority in the discovery of thorium radioactivity.

Always in Germany, Julius Elster (1854–1920) and Hans Friedrich Geitel $(1855–1923)$, an inseparable couple of teachers at the Wolfenbüttel Schule, known essentially for having discovered the photoelectric cell, proved experimentally at the end of 1898 that the radiation emitted by uranium salts was the same at ambient pressure, under vacuum, and in a mine at a depth of 853 m. In 1901, they measured the radioactivity of the air and soon after that of the ground and studied the radioactive decay for long time. In the 1902–1905 period, Friedrich Oskar (Fritz) Giesel (1852–1927) from the Braunschweig University observed the presence of a new radioactive substance in minerals of lanthanum and cerium that he named emanium since it emitted a radioactive gas (Giesel [1900–1905](#page-29-0)). Soon, however, it became clear that the emanium was nothing other than actinium, previously discovered by Debierne.

In 1900, Becquerel, by precipitation of insoluble carbonates from a solution containing uranyl ions, discovered the strange fact that the uranium was left in solution in the form of a soluble uranyl carbonate but that most of the radioactivity disappeared from the solution, appearing instead in the precipitate that did not contain uranium. There was therefore some radioactive material that "emaned" from the uranium salts and that escaped from the solution with the precipitate (Becquerel [1901\)](#page-28-0).

In 1900, while investigating the radiochemical properties of uranium, W. Crookes and Becquerel made an important discovery: by precipitation of a carbonate salt from a solution containing uranium, they discovered that while the uranium remained in the supernatant as a soluble uranyl carbonate complex, the radioactivity originally associated with the uranium was now present in the precipitate, which contained no uranium. Moreover, the radioactivity of the precipitate slowly decreased with time, while the supernatant showed a corresponding increase of radioactivity. Similar results were obtained by E. Rutherford and F. Soddy when investigating the radioactivity of thorium.

The existence of radioactive gases that seemed to be present in receivers "emanating" from radioactive materials added further complications to the research involved in arranging the newly discovered elements in the periodic table (Curie [1899](#page-28-0)). In 1899, Robert Bowie Owens (1870–1940) (Owens [1899](#page-30-0)) and Ernest Rutherford [\(1900a](#page-30-0), [b\)](#page-30-0) identified an emanation of thorium, and in the same year Pierre and Marie Curie observed that even radium produced an emanation that, in contrast to that of thorium, remained radioactive for months (Curie and Curie [1899\)](#page-28-0). The occurrence of thorium and uranium emanations was further confirmed by Friedrich Ernst Dorn (1848–1916), professor at the Friedrichs Universität of Halle (Dorn [1900\)](#page-29-0).

In 1900, William Crookes, who in 1861 had isolated thallium, succeeded in isolating at the same time as Becquerel a uranium fraction that seemed to possess the full emission. He named this fraction of uranium as uranium-X (later identified as protactinium). He also observed that the radioactivity of uranium-X decayed very quickly while in the meantime the radioactivity of the original uranium was restored. In 1903, A. Debierne discovered the same emanation in actinium (Debierne [1903](#page-29-0)). In 1909, Daniel Strömholm (1871–1961), professor of chemistry at the University of Uppsala, and Theodor Svedberg (1884–1971), at that time still associate professor, used isomorphism to identify the chemical character of the new radio elements. By crystallization of different salts in solutions containing radio elements, they found that thorium X (^{208}Pb) crystallized together with the lead and barium salts, but not with other salts, and concluded that it was an alkaline earth substance in contrast to previous erroneous conclusions that it was a univalent element.

In 1904, William Ramsay and John Norman Collie (1859–1942) at University College London suggested eliminating the term emanation from the terminology of the radioactive compounds and using the prefix eka, initiated by Mendeleev, for the unknown elements of the periodic table. In this way the emanations would be called eka-thorium, eka-radium, eka-actinium, etc. Their proposal, however, fell on deaf ears. In 1910, Ramsay went back to the problem together with Robert Whytlaw-Gray (1877–1958), suggesting (Ramsay and Whytlaw-Gray [1910\)](#page-30-0) the name niton for the radium emanation. Despite these repeated attempts, the term emanation survived until 1923 when the international commission for the chemical elements chose the name radon, for all emanations (Aston et al. [1923](#page-28-0)). The different emanations, in fact, all possessed the same absorption spectrum that resembled that of the rare gases, argon, krypton, and xenon, a fact that suggested that it was an element of the family of the rare gases. This new element was isolated and studied by Rutherford and Soddy in 1902 and by Ramsay and Whytlaw Gray (1877–1958) in 1908. In few years it was clear that the different emanations were all produced by the radioactive decay of various isotopes of the elements. Radon was found very useful in medicine, especially in cancer research and in industrial tests, and was even utilized in the control of industrial welding processes.

6.4 Transmutation of the Elements

At the beginning of the twentieth century, radioactivity appeared as a new chapter of inorganic chemistry, relative to a small group of rather exotic elements, rarely found in nature, and barely utilizable in industry, with at best some useful applications for medical treatments and for dating archeological objects. For the physicists, radioactivity continued to be a mysterious topic since the nature of the emitted rays was still unknown and nobody knew its relationship with atomic structure. The scientist that did realize the connection between atomic structure and radioactivity within few years, and paved the way to the study of the nuclear structure, was a New Zealander, Ernest Rutherford, another great representative of the school of physics that, from Lord Kelvin to John Thomson, had made Cambridge the center of the physical world of that period. Ernest Rutherford, whose research on the atomic structure are discussed in Chap. [5](http://dx.doi.org/10.1007/978-3-642-28180-8_5), started his studies on radioactivity when in 1896, as a young researcher at Cambridge in the laboratory directed by J. J. Thomson, he assisted in the spread of the news of the discovery of Becquerel's U rays. The unknown nature of these rays stimulated his interest and pushed him to investigate their composition. In 1898, he succeeded in showing that the U rays were made of two types of particles that he named α and β rays, different for their penetrating power into matter, as well as for their charge and for their mass. The α rays were absorbed by thin metallic sheets of a few millimeters thickness while the β rays were able to cross metallic sheets 100 times thicker. The paper, published in 1899 when Rutherford was at the McGill University in Canada (Rutherford [1899\)](#page-30-0), was sent by J.J. Thomson to the editor of the "Philosophical

Magazine" on 1 September 1898, almost contemporaneously to that of Marie Curie on radioactivity. In 1899, Fritz Geisel, Antoine-Henri Becquerel, and Marie Curie proved that the β rays were very fast electrons. The nature of the α particles, however, was understood only after some years. In the period 1906–1912, the German Hans Geiger (1882–1945) moved to Manchester to work with Rutherford who was now professor of physics there. In 1908, Rutherford and Geiger built an instrument to measure and count α particles and proposed that these were He nuclei (Rutherford and Geiger [1908\)](#page-30-0). In 1909, Rutherford and Thomas Royds (1884–1955) definitively confirmed this hypothesis (Rutherford and Royds [1909](#page-31-0)).

The emission of new rays from radioactive materials continued to present new surprises; in 1900 the French chemical-physicist Paul Villard (1860–1934), working at the École Normale Superieure of Paris, discovered that uranium compounds emitted a third type of radiation even more penetrating than x-rays, able to get through a lead plate of several centimeters thickness (Villard [1900\)](#page-31-0). Villard proved that these rays, that he named γ rays, were not deviated by an electric or magnetic field and therefore concluded that they were of the same nature as x-rays.

Rutherford's research activity was recognized as being of the highest level by everybody, and in particular by Thomson, his principal supporter. The rules of the University of Cambridge were, however, extremely rigid and offered few hopes to obtain in a short time a stable position for him. An interesting opportunity occurred, however, when a professorship opened at the McDonald Laboratory of the McGill University of Montreal in Canada. Rutherford decided to try this adventure and in 1898 left England for the new seat. The laboratory had been founded in 1891 thanks to the financial support of a philanthropist, Sir William MacDonald, who also supplied the funds to establish there the chair of physics that Rutherford occupied.

The McGill laboratories were well equipped and had one of the best collections in the world of radioactive materials. Rutherford therefore had no difficulties in resuming the work that he had started at Cambridge. Once arrived at the McGill in 1899, he started to collaborate with Robert Bowie Owens (1870–1940), professor of electro-technics at the same university, who studied the ionizing power of the thorium radiation. Owens had started his research with a normal sample of thorium, but the results of his measurements seemed completely erratic, subject to inexplicable sudden variations depending on the strangest factors such as the opening of a door of the laboratory or the displacement of a person around the working table.

After some time, Owens was convinced that the measurements were completely untreatable and that the results were random, due to air currents. For instance, if the system was placed in a closed receiver in which air circulated, the radioactivity decreased but if it was left quiet for about a quarter of an hour, the radioactivity went back to the initial value. Owens, bored and annoyed, abandoned the problem in the hands of Rutherford who, in a relatively short time, was able to prove that the radioactivity was not induced in the air around the sample, but was an emanation escaping from the thorium oxide in the form of a radioactive gas capable of making radioactive all materials that it contacted (Rutherford [1900a,](#page-30-0) [b\)](#page-30-0).

The phenomena exhibited by thorium compounds receives a complete explanation if we suppose that, in addition to the ordinary radiation, a large number of radio-active particles are given out from the mass of the active substance. This "emanation" can pass through considerable thicknesses of paper. The radio-active particles emitted by the thorium compounds gradually diffuse through the gas in its neighbourhood and become centres of ionization throughout the gas.

Furthermore, by plotting in a diagram the variation of the radioactivity intensity as a function of time, he obtained an exponential curve that allowed him to define the concept of half-life, i.e., the time necessary to reduce to one half the radioactivity of a sample. His first Canadian student, Harriet Brookes (1876–1933), who became the first female nuclear physicist in Canada, collaborated with him on the series of research on the thorium emanation.

In the years 1900–1901, Rutherford continued to study thorium and radium $(^{226}$ Ra) emanations. In 1900, a young English chemist, Frederick Soddy (1877–1956), coming from the University of Oxford, where he had worked as researcher from 1898 to 1900, joined Rutherford's research group at the McGill University to work with him on thorium radioactivity. The collaboration with Soddy gave rise to an intense activity in analytical chemistry to identify the unknown atoms which appeared and disappeared in connection with the thorium emanation. With reference to this period, Rutherford used to tell his friends that the fastest transformation he knew was his own from a physicist to a chemist in these days!

Their work with microscopic quantities of substances which changed over time, without supplying the smallest hint of what was happening, led them to look for models in order to find the right direction to follow. They continued formulating new conjectures that, after a short while, were found to be wrong. For 2 years they collected a large amount of data and invented an equivalent number of fanciful interpretations of the thorium emission, trying to understand how they collected the energy necessary to cross successive layers of aluminum sheets.

In 1902, Rutherford and Soddy started also a research on the emission of thorium using a new sample of pure thorium nitrate arrived from Germany. Soddy decided to prepare thorium hydroxide from the nitrate by precipitation with sodium carbonate or ammonium hydroxide. By filtration he collected in the filter solid thorium hydroxide that had completely lost its radioactivity. By evaporation of the filtrate he found, however, a solid residue which was strongly radioactive. They named thorium-X this new radioactive material as done by Crookes who named uranium-X the emission of uranium. In January 1902, after the Christmas holidays, they discovered in a new experiment that thorium-X was produced from thorium at a constant speed and that it decayed following an exponential law. Rutherford and Soddy hurried to publish these results in two papers in which they clearly pointed out that a new type of matter was produced in a kind of chemical reaction that occurred inside the nucleus (Rutherford and Soddy [1902a,](#page-30-0) [b](#page-30-0)):

The position is thus reached that radioactivity is at once an atomic phenomenon and the accompaniment of a chemical change in which new kinds of matter are produced. The two considerations force us to the conclusion that radioactivity is a manifestation of subatomic chemical change. [...]The idea of the chemical atom in certain cases spontaneously breaking up with the evolution of energy is not of itself contrary to anything that is known of the properties of the atoms, for the causes that bring about the disruption are not among those that are yet under our control.

In order to spread this information throughout the physics community, they published the same article in the Philosophical Magazine, a typical physics journal, repeating with different words the same concepts (Rutherford and Soddy [1902b](#page-30-0)); after 2,000 years Rutherford had discovered that the alchemist's dream, the transmutation of the elements, was a reality and existed in nature! These conclusions were so important and innovative that Rutherford hurriedly wrote to Crookes, editor of the Journal of the Chemical Society, requesting him to ensure that publication would not suffer any delays.

Soddy and Rutherford continued their collaboration trying to measure the energy released in the radioactive decay. In February 1903, Rutherford found that the α particles emitted in the decay were heavy particles carrying a positive charge and obtained a ratio e/m between the charge and the mass of 6,000 with respect to the value 10,000 of e/m for hydrogen, concluding that about 99% of the energy was carried by the α particles. In three successive 1902 papers, they repeatedly pointed out that in all minerals containing uranium, helium always appears as an inclusion, a fact that encourages the supposition that it is a final product of the radioactive decay.

In 1903, Soddy left Canada to go to work with William Ramsay at University College London, where he continued his research on radium emanation. In the same year, Soddy and Ramsay published a paper in which they announced that, using emission spectroscopy, they had discovered that the helium was produced in the radioactive decay of radium bromide as well as in the decay of the emanation (Ramsay and Soddy [1903\)](#page-30-0).

In 1904, Soddy took up an appointment at Glasgow's University where he stayed for 10 years until in 1914 he became professor of chemistry at the University of Aberdeen. At Glasgow, Soddy discovered the existence of the isotopes and developed his displacement law that establishes the rule governing the transmutation of an element during radioactive decay. According to this law, better known as the Fayans and Soddy law, a radioactive element emitting an α particle shifts back two places in the periodic table while it moves forward one place by emitting a β particle or back one place by capturing an electron. This law represented the rule for the construction of the radioactive families of elements (Soddy [1913\)](#page-31-0).

The term "isotope" was suggested to Soddy in 1913 by Margaret Todd, a Scottish doctor and writer to whom he was distantly related. Talking to her, Soddy tried to explain that, according to his research, it did look like different elements occupied the same place in the periodic table. The learned Todd noticed immediately that in Greek "the same place" is called σ - σ σ ₀ σ ₅ (*iso-topos*) and suggested that he use it. Soddy accepted the suggestion right away and since then the term isotope has been in the public domain.

For his investigations of the disintegration of the elements and of the chemistry of radioactive substances, and in particular for the discovery of the displacement law and for the concept of isotope, Soddy received the Nobel Prize for chemistry in 1921.

Almost at the same time the Pole Kazimierz Fajans (1887–1975), in collaboration with Otto Göhrling, studied the sequence of the radioactive transformations of uranium 238 U decay (Fajans [1913a,](#page-29-0) [b](#page-29-0)) and also arrived at the idea of the existence of isotopes. Fajans and Göhrling actually realized, using the idea of the radioactive displacements, that by emission of an α particle followed by emission of β and γ rays, uranium 238 transformed into uranium 234 according to the nuclear reaction

$$
^{238}\text{U} \stackrel{\alpha}{\longrightarrow} ^{234}\text{Th} \stackrel{\beta,\gamma}{\longrightarrow} ^{234}\text{Pa} \stackrel{\beta,\gamma}{\longrightarrow} ^{238}\text{U}
$$

They called the unstable nucleotide 234 Pa of protactinium of very short half-life (77 s) brevium (from the Latin brevis). The following year, 1914, a paper appeared written by Theodore William Richards, professor at Harvard and future Nobel laureate for his very accurate atomic weights measurements, and by Max Lembert, a student of Fajans who had gone to work with Richards. They reported several different atomic weights for lead extracted from different samples (Richards and Lembert [1914](#page-30-0)). In this paper, the authors pointed out the agreement of their results with the hypothesis of the existence of isotopes of Fajans and Soddy:

This amazing outcome is contrary to Harvard experience with several other elements, notably copper, silver, iron, sodium, and chlorine, each of which seems to give a constant atomic weight, no matter what the geographical source may have been. No attempt is made here to discuss the theoretical aspects of the facts presented, but attention is called to their qualitative agreement with the hypothesis of Fajans and of Soddy.

The existence of protactinium was confirmed after few years in 1918 when Otto Hahn and Lise Meitner in Germany and Frederick Soddy and John Cranston (1891–1972) in England independently identified the isotope 231 Pa. In 1934, Aristide von Grosse (1905–1985) from the University of Chicago succeeded in preparing 2 mg of the oxide and 7 years later he obtained the pure metal (von Grosse [1934\)](#page-29-0).

Kazimierz Fajans, after having studied at Leipzig, Heidelberg, and Zurich, went to Manchester in 1910 to the laboratory directed by Rutherford where he worked in collaboration with Moseley. Later he returned to Germany, first to the university of Karlsruhe and then as professor of chemical physics to Munich. In 1935, with the growth of the Nazi persecution of the Jews, he left Germany and immigrated to the United States as professor at the Ann Arbor University in Michigan. In collaboration with Otto Hahn ([1936\)](#page-29-0) he discovered the conditions of precipitation and absorption of radioactive substances, an important technique of separation and cleaning of small traces of radioactive substances. In 1924, a solid candidature of Fajans was submitted to the Nobel Prize committee. The great majority of the scientific community involved in radioactive activities was almost certain that he would get it, up to the point that the Swedish magazine "Svenska Dagbladet" announced his victory the day before the final decision of the Nobel committee. Unfortunately for him, the next day the Committee announced that no prize, either

in chemistry or in physics, was to be awarded that year. After this he was again proposed twice but without success.

The proof of the existence of isotopes came later from the J.J. Thomson group, in particular from Thomson's collaboration with Francis William Aston which presented experimental evidence for the existence of two isotopes of neon (Thomson [1913a](#page-31-0), [b](#page-31-0)).

At the beginning of the twentieth century, about 40 radioactive elements were already known and among them several had exactly the same chemical properties although a different atomic weight. For example, ten different varieties of thorium with atomic weights ranging from 232 to 212, two of uranium, and six of actinium had already been identified and there was no place in the periodic table to allocate this multitude of elements. This fact made several chemists suspicious that one should completely reexamine the periodic table, one of the milestone of the structure of chemical theory, since in the table there were only six places available to locate a much larger number of elements, between the position of hydrogen and that of uranium, corresponding to positions 43, 61, 72, 75, 85, and 87.

In 1907, Thomson started his research on the canal rays, made of positively charged particles. Letting a jet of neon atoms cross an electric and a magnetic field, he registered on a photographic plate two traces, corresponding to two types of particles, one with atomic weight 20 and the other 22. To explain this strange result Thomson proposed the existence of either an unknown neon compound, for instance, a neon hydride, or of a new element, meta-neon. The occurrence of two traces did not seem, however, too convincing for the chemistry community without a true chemical separation of the two types of particles and was considered by many as an experimental artifact.

In 1909, Francis William Aston (1887–1945), who started to be interested in cathode rays at Birmingham under the guide of the chemists William Augustus Tilden (1842–1926) and Percy Frankland (1858–1946) and of the physicist John Poynting (1852–1914), went to work at the Cavendish Laboratory upon the invitation of Thomson to assist him in his experiments on the canal rays. Thomson committed Aston first to the problem of improving the equipment that had been built and then to the study of the existence of this strange meta-neon.

Aston improved Thomson's apparatus and in 1913 with the new instrument Thomson and Aston again studied neon and announced that the element of mass 22 had the same properties as the neon of mass 20. After having identified spectroscopically the two isotopes of neon, Aston dedicated himself to their separation. First he tried without success the fractioning on coal cooled in liquid air. In 1913, he used with more success gas diffusion through tubes of porous clay, using a quartz microbalance to control the different stages of the separation procedure of the two species.

With the start of the First World War in 1914, Aston was forced to stop his research and, because of his competence in mechanics and combustion processes, was assigned as technical assistant to the Royal Air Force factory at Farnborough to study the effects of atmospheric conditions on materials for aeronautics.

Back at the Cavendish Laboratory in 1919, he built a new instrument much more efficient than the first. In Thomson's original apparatus, the electric and magnetic fields were superposed so that the particles crossed the two fields simultaneously. However, Aston separated them, letting the magnetic field act on the particles already separated by the electric field. Using several ingenious devices, Aston succeeded in focusing at the same point of the photographic plate all particles possessing the same charge/mass ratio. With this new instrument, Aston definitively proved the existence of two neon isotopes. Soon after, he discovered that even chlorine had two isotopes 35 Cl and 37 Cl (Aston [1920b](#page-28-0)) and, having understood that the existence of the isotopes was a general feature of matter, started a systematic study of the isotopic effect, discovering that bromine, lithium, boron, and argon also had two isotopes, that magnesium had three, krypton and sulfur six, and xenon seven. In the following 3 years, he analyzed more than 30 elements and in total in his full research activity he identified 212 isotopes of different elements. In 1921, he decided to build a new instrument with much better performances than the previous one, overall with a better resolving power, at the point that he succeeded in separating 6 isotopes of mercury in the range from 198 to 204 atomic masses. Aston summarized the results of his research in two famous papers (Aston [1919](#page-28-0), [1920a](#page-28-0)) and in the book Isotopes of 1922 (Aston [1922](#page-28-0)).

In the introduction to the book he wrote

The importance, from purely practical and technical points of view, of the theory of isotopes would have been insignificant had its application been confined to the radioactive elements and their products, which are present in infinitesimal quantities on the Earth. But now that the isotopic nature of many elements in everyday use has been demonstrated, the possibility of their separation to any reasonable extent raises questions of the most profound importance to applied science.

For this research, he was awarded the Nobel Prize for chemistry in 1922 "for his investigations into the disintegration of the elements, and the chemistry of radioactive substances." In 1937, Aston built a third prototype, far more advanced than the previous one, with a resolving power about 20 times better and a precision in the determination of mass about 100 times greater (Aston [1937](#page-28-0)).

The apparatus built by Aston was called a mass spectrograph since it allowed one to separate ions or neutral atoms of different atomic mass. With this technique, it was easy to characterize the isotopes of even well-known elements. For instance, William Francis Giauque (1895–1982) proved the existence of three isotopes of oxygen of atomic mass 16, 17, and 18 (Giauque and Johnston [1929a](#page-29-0), [b](#page-29-0)) and Harold Clayton Urey discovered deuterium (Urey et al. [1932a](#page-31-0), [b](#page-31-0)), the isotope of hydrogen of atomic mass 2.

Aston's mass spectrograph has been perfected over the years and represents today an analytical tool of fundamental importance in all fields of chemical, biological, and pharmaceutical research.

6.5 Completion of the Periodic Table

The discovery of so many new elements made it necessary to modernize the periodic table, introducing new classificatory techniques more in line with the recent theoretical developments. By now it was clear that atomic weight was too coarse a variable for correct positioning of the elements in the table. This concept had been already expressed in 1886 by Johannes Rydberg (1854–1919), the first to mention that the order number of the elements was the only independent variable that could be expressed by integer numbers (Rydberg [1886\)](#page-31-0). In addition, new elements, such as polonium, actinium, radium, radon, and the whole series of different isotopes, had been discovered and many of them were not easily inserted into the Mendeleev table.

In 1911, a Dutch lawyer and amateur physicist, Antonius Johannes Van den Broek (1870–1926), suggested that the number representing the position of an element in the periodic table, later the atomic number Z, corresponded to the total number of electrons in the atom. This idea was presented in a paper that Van den Broek published in the journal Nature (Van den Broek [1911\)](#page-31-0) in July 1911, only 1 month after the publication of the famous article of Rutherford on the atomic nucleus. In two successive papers, (Van den Broek [1913a](#page-31-0), [b\)](#page-31-0) van den Broek maintained that it was an error to compare the nuclear charge to the atomic weight of an element, suggesting that instead one should use the atomic number which represents the number of positive charges on the nucleus as proved by the experiments of diffusion of the α particles.

Rutherford had limited his discussion in his paper to the observation that the nuclear charge was an integer number of the order of half of the atomic weight, starting from the assumption that the atomic nuclei were made of helium nuclei, each with a nuclear charge equal to one half of its atomic weight. This implicitly corresponded to supposing that the nuclear charge was equal to the atomic number.

Henry Gwyn Jeffreys Moseley (1887–1915) strongly contributed to the final arrangement of the periodic table. He was one of the most brilliant pupils of Rutherford, prematurely killed in action during the First World War at the age of 27 years in the battle of Gallipoli in Turkey on 10 August 1915, being hit in the head by a sniper while, as signal officer of the 38th brigade of the Royal Engineers, he was calling headquarters on the phone. Son of a professor of anatomy at Oxford and a student of Trinity College of the same university, Moseley went to work with Rutherford in 1910 when the latter was professor at Manchester. In 1913 he discovered the law that bears his name, a systematic relationship between the frequency of the X-rays and the position of the elements in the periodic table. In particular, he found that a perfect linear relation is found by plotting the square root of the x-ray frequency as a function of atomic number (Moseley [1913](#page-30-0), [1914\)](#page-30-0).

Up to that time the concept of atomic number defining the position of the element in the periodic table was considered only representative of the sequence of the atomic mass and possessed no direct structural meaning. Moseley's law showed instead that the atomic number was a perfectly measurable quantity, confirming the hypothesis of Van den Broek in that it represented the number of electrons in the atom and thus the number of positive charges in the nucleus.

Moseley's measurements allowed one to define the exact position of each element in the periodic table, solving many still unanswered questions. For example, in building his table on the basis of chemical properties, Mendeleev had inverted the position of cobalt with that of nickel, locating cobalt at position 27 and nickel at position 28, despite the fact that the atomic weight of cobalt was slightly higher than that of nickel. Moseley's data perfectly confirmed this genial intuition of Mendeleev. Furthermore, Moseley showed that in the atomic numbers sequence, the positions 43, 61, 72, 75, 85, and 87 were still empty, confirming on one hand Mendeleev's prediction and supplying on the other the key to localize exactly the missing elements.

The existence of the radioactive element 61, promethium, had been foreseen in 1902 by the Czeck chemist Bohuslav Brauner (1855–1935), professor of inorganic chemistry at the Carl IV University of Prague. Brauner was one of the principal experts on rare earths. He anticipated the existence of the isotopes and suggested the use of oxygen as a basis for the atomic weights of the elements. A good friend of Mendeleev, he investigated the position of the rare earths in the periodic table (Brauner [1901\)](#page-28-0) that he extended alongside lanthanum, suggesting that all rare earths should be placed in a single position of the periodic table between lanthanum (57) and tantalum (73). Brauner's proposal was not accepted due to the opposition of Mendeleev, until Moseley confirmed beyond any doubt that only 14 elements existed after lanthanum. This gave rise to the birth of the lanthanide series, from lanthanum (57) to lutetium (71), forming a small separate periodic table. Actually the number of lanthanides was an open problem for chemistry at the beginning of the twentieth century, since for several of them the pure products did not exist in appreciable quantity, being difficult to separate chemically mixtures of very similar elements. It was only thanks to Moseley's data that the problem was definitively solved.

While working on the rare earths, Brauner realized that the difference between neodymium and samarium was too big in comparison to the other lanthanides and made the hypothesis of the existence of a new element at position 61 between them (Brauner [1926\)](#page-28-0). Different research groups all over the world declared within a few years to have somehow identified element 61, without, however, being able to prove it. The discovery of new elements had actually become at the end of the nineteenth and the beginning of the twentieth century quite a mass phenomenon. A certain Kosmann, unknown as a scientist, even reported in 1896 in a chemical journal a kind of April Fool joke, saying that he had succeeded in detecting two new elements at once, with the pompous names of kosmium and neokosmium. It soon became apparent that the "discoverer" of kosmium and neokosmium had just been making fun of this discovery's popularity.

The two most famous cases of imaginative self-deception were those of the hypothetical elements *florentium* and *illinium*. In 1924, Luigi Rolla (1882–1960), professor at the University of Florence, Italy, insisted that he had discovered, in collaboration with his student Lorenzo Fernandes (1902–1977), element 61 and sent a letter to the Accademia Nazionale dei Lincei containing the result of his spectroscopic analysis (Rolla and Fernandes [1926](#page-30-0)). Two years later in 1926 at the

University of Illinois, Urbana, B. Smith Hopkins (1873–1952), an expert on the chemistry of the rare earths, and his coworkers Leonard Yntema and J. Allen Harris, maintained that they had also identified element 61 and proposed for it the name illinium (Hopkins et al. [1926](#page-30-0)). All attempts made by experts in this field, such as Wilhelm Prandtl as well as the Noddack husband and wife, to confirm the validity of the experimental arguments supplied by the two group for florentium and illinium, respectively, resulted in inconsistencies because it was soon clear that it should be a radioactive element. The proof of the existence of element 61 was only obtained in 1945 by Charles D. Coryell (1912–1971), chief of the section for the study of the products of nuclear fission of the Manhattan project and his coworkers Jacob Akiba Marinsky (1918–2005) and Lawrence E. Glendenin (1918–2008) during the analysis of some by- products of uranium fission, made using an ion exchange column. The presence of the isotope of mass 147 of element 61 was confirmed using mass spectroscopy.

Due to the secrecy that surrounded all research connected with the war, the discovery was only announced in 1947. Coryell, acting upon the suggestion of his wife, proposed the name *prometheus*. In 1949, the International Union for Chemistry decided on the present name of promethium.

Mendeleev had already predicted the existence of element 75 with chemical properties similar to those of manganese that he named eka-manganese. In 1925, two researchers from Berlin, Walter Noddack and Ida Tackle, who married in 1926, after having searched for long time for eka-manganese in different minerals such as columbite, gadolinite, molybdenite, and several minerals of platinum, published a paper (Noddack et al. [1925](#page-30-0)) declaring that they had identified, with the help of the x-ray expert Otto Berg from the Siemens laboratories, element 75 that they called rhenium in honor of the Rhine river. Only in 1927, however, did they succeed in obtaining 1 g of rhenium from more than 600 kg of molybdenite (Noddack and Noddack [1927\)](#page-30-0). The couple also claimed to have identified the element of atomic number 43 that they called *masurium* in honor of the Masuria, region of Eastern Prussia where Walter Noddack was born, but they never succeeded in proving experimentally that they really had found it. Element 43 was instead identified in 1937 at the Physics Institute of the University of Palermo, Italy, by the mineralogist Carlo Perrier (1886–1948) and the physicist Emilio Segre` (1905–1989). It was called *technetium* from the Greek τ_{E} to τ_{E} , artificial, because it was artificially produced (Perrier and Segrè [1937](#page-30-0)). Segrè had brought from Berkeley to Palermo a sample of molybdenum obtained from Ernest Lawrence with whom he collaborated. The molybdenum was bombarded with deuterium nuclei at the Berkeley cyclotron, creating the technetium isotopes $95Tc$ and $97Tc$. Technetium was thus the first artificially created element.

The element 72, hafnium (from *Hafnia*, the Latin name of Copenhagen) was discovered in 1923 by the Hungarian George Charles de Hevesy and by the Dane Dirk Coster at Copenhagen, the city where both worked with Niels Bohr.

The most credited version of the discovery of hafnium is based on the belief that the chemists of the time thought that element 72 had to be a rare earth. Apparently Niels Bohr, analyzing in 1921 the electron distribution of the electrons in the atomic levels, realized instead that with lutetium the fourth electronic shell was completed and therefore that element 72 could not be a rare earth. It is therefore said that Bohr asked his coworkers de Hevesy and Coster to look for it in the zirconium minerals, a choice that allowed them to find it in a relatively short time, and that Carl Popper utilized it to confirm his theory of the reduction of chemistry to the quantum theory. This version, however, has been recently disputed by Eric R. Scerri of the department of chemistry and biochemistry of the University of California, Los Angeles, and editor of the journal Foundations of Chemistry (Springer) (Scerri [1994](#page-31-0)). Scerri bases his different version on a series of documents obtained from the son of Friedrich (Fritz) Paneth, showing that it was not Bohr but the same Paneth, with whom de Hevesy had collaborated at the radium institute at Vienna, who suggested looking for hafnium in zirconium minerals. Friedrich Adolf Paneth (1887–1958), in addition to being an expert in radiochemistry, was also competent with epistemological problems of chemistry and published several papers on the topic, in particular a manuscript of more than 200 pages of a lecture that he gave at Konigsberg in honor of Kant, titled Die erkenntnistheoretische Stellung des chemischen Elementbegriffs. This lecture, published in German in the records of the Königsberger Gelehrten Gesellschaft, was later translated into English by his son with the title *The epistemological status of the chemical concept of element* (Paneth [1962\)](#page-30-0). Paneth was an antireductionist, convinced that the network of empirical relationships created by the great chemistry of the nineteenth century could not be replaced by mathematical techniques, as he pointed out in his lecture:

Even if the character of chemistry should change essentially in the future owing to penetration by mathematico-physical methods, its history during the nineteenth century, in which it achieved such successes without mathematics, must never be ignored in its philosophic evaluation.

This could explain why the suggestion of Paneth to de Hevesy was based on chemical knowledge and not on electronic configuration.

Element 87 was the last to be discovered, even if there were clear hints that it should be chemically similar to the alkaline metals. Mendeleev had in fact called it eka-cesium, and, in addition, it was expected by experts that it would be radioactive, being intermediate between two radioactive elements, radon and radium.

Despite the fact that few doubts existed on its radioactive nature, several attempts were made to identify a stable eka-cesium, obviously without results, except that of filling the literature with fanciful names such as *russium* (1925), alkalinium (1929), virginium (1932), moldavium (1937), mosandium (named after the Swedish chemist, Mosander), etc. With the publication of the law of the radioactive displacement of Moseley it was clear that element 87 could either be derived from actinium 89 by emission of an α particle or from radon by emission of a β electron:

$$
{}^{227}_{89}\text{Ac} \longrightarrow {}^{223}_{87}\text{Ac} + {}^{4}_{2}\text{He}
$$

$$
{}^{222}_{86}\text{Rn} \longrightarrow {}^{222}_{87}\text{eka} - \text{Cs}
$$

and therefore research on element 87 was essentially limited to these two possible nuclear reactions, preferentially relative to the actinium decay since there were serious doubts about the possibility of a β emission from radon.

At the end of the 1930s, the Institute du Radium of Paris was strongly involved in the study of actinium, discovered in 1900 by Andre´ Louis Debierne, the director of the institute, where also worked Irène, daughter of Marie Curie and wife of Jean Joliot. Debierne had a bad character and was by no means a good director. He did not get along too well with Irène Joliot-Curie to the point where they worked on the same problem without even informing each other of their progress. At their service worked a young technician, Marguerite Perey (1909–1975), appointed by Marie Curie in 1929 who had learned from her the techniques of purification and handling of radioactive materials.

Debierne and Irène Curie both separately asked Marguerite Perey to purify actinium samples. Irène Joliot-Curie was interested in determining the half-life of the 227 (^{227}Ac) nucleotide whereas Debierne on his side was interested in looking for new, not better identified, radio elements (Adloff and Kauffman [2005\)](#page-28-0).

Working on the purification of actinium, Perey discovered that the 227 actinium could decay either by emission of a β electron, thereby giving rise to thorium 227, or by emitting an α particle and producing a new element according to the scheme

This new element, temporarily called *catium*, produced by emission of a β electron the 223 isotope of radium.

Marguerite Perey separately informed Debierne and Irène Curie of her experiment with the expected result that each of the two contenders considered themselves as the true inspirer of Perey's research. In particular, when Irène Curie went to see Debierne and told him that the technician Perey discovered a new element during research that she, Irene Curie, had directed, a violent fight started between them so that at the end they both decided not to be associated with the research that was therefore published under the name of only Marguerite Perey [\(1939](#page-30-0)). The name catium selected by Perey did not, however, please Irène Curie or the other members of the institute, and was replaced by the name francium, obviously acceptable to everybody.

The development of nuclear chemistry has strongly benefited from the availability of projectiles such as α particles and from the discovery of a new nuclear particle, the neutron. In 1919, Rutherford, striking nitrogen atoms with α particles, realized for the first time in the laboratory the alchemist's dream of transforming one chemical element into another (Rutherford [1920\)](#page-31-0):

$$
{}_{2}^{4}\text{He}^{++} + {}_{7}^{14}\text{N} \rightarrow {}_{8}^{17}\text{O}^{+} + {}_{1}^{1}\text{H}^{+}
$$

In this nuclear reaction, he obtained the emission of ionized hydrogen atoms (protons) and realized that the protons were part of the structure of the atomic nucleus:

We must conclude that the nitrogen atom is disintegrated under the intense forces developed in a close collision with a swift alpha particle, and that the hydrogen atom which is liberated formed a constituent part of the nitrogen nucleus.

The term proton was probably coined just by Rutherford since, according to Pais [\(1991](#page-30-0)), it appeared for the first time in Rutherford's paper Nuclear Constitution of the Atoms (Rutherford [1920\)](#page-31-0).

In 1920, Rutherford knew only two types of particles, protons and electrons, and with only these two particles it was problematic to build a theory of nuclear structure that could satisfy the laws of physics and at the same time account for the fact that the mass of the atom was roughly double that of the nuclear charge. There was another problem to explain, that of the emission of the B rays, electrons that undoubtedly came from the nucleus. If electrons were present in the nucleus as true particles, they had to travel around the protons on orbits of extremely small dimensions, this implying that they possessed monstrously huge energies. For example, the α particles, that were helium nuclei, should have been made of four protons bound together by two electrons. The impossibility of this kind of nuclear structure was theoretically proved in the 1930s by the Russian Armenian astrophysicist Viktor Ambarzumian (Hambardzumyan) and by the Ukrainian theoretical physicist Dmitri Iwanenko (Ambarzumian and Iwanenko [1930a](#page-28-0), [b](#page-28-0); Iwanenko [1932\)](#page-30-0).

Even more difficult to explain was the nuclear structure of heavy elements like uranium, for which hundreds of electrons had to be contained in the extremely small volume of the nucleus. Rutherford therefore made the hypothesis of the existence of a third type of elementary particle, electrically neutral, in which in an unknown way a proton and an electron were fused together. He called this unknown particle a neutron from the Latin root *neutral* with the Greek ending on in order to find the correct assonance with the names of the known elementary particles proton and electron. Of course all students of the Cavendish Laboratory were immediately addressed to look for this new particle without, however, any significant result.

A new series of research opened the road to the discovery of the neutron. In 1930, Walther Bothe, one of the great German nuclear physicists, Nobel Prize winner for physics in 1954, and his coworker Herbert Becker discovered that beryllium ($_4^{9}$ Be) bombarded with α particles emitted a new type of highly penetra-
tive radiation able to cross up to 20 cm of lead 200 times more efficient than tive radiation able to cross up to 20 cm of lead, 200 times more efficient than protons, and concluded that it was made of very high-energy γ rays.

In 1932, Irène Joliot-Curie and her husband Jean Joliot published a paper entitled E mission de protons de grande vitesse par les substances hydrogénées sous l'influence des rayons γ trés pénétrants (Curie and Joliot [1932\)](#page-29-0) in which they maintained that by bombarding paraffin or other substances containing hydrogen with these γ rays emitted from beryllium, one obtained the emission of very high-speed protons (De Gregorio [2006](#page-29-0)):

Nous avons étudié ces rayonnements par l'ionisation qu'ils produisent dans une chambre montée sur un èlectromètre Hoffmann.[...] Le courant augmente notablement quand on interpose des écrans de substances contenant de l'hydrogène comme la paraffine, l'eau, le cellophane. L'effet le plus intense a été observé avec la paraffine[...] Ces rayons γ de
grande énergie sont capables de projeter des protons de grande vitesse quand ils traversent une substance hydrogénée.[...] Cet effet s'accompagne d'une absorption considérable du rayonnement par les noyaux d'hydrogène.

In the meantime, an old pupil of Rutherford at Manchester, James Chadwick, who in 1913 had left England to work in Berlin with Hans Geiger, went back in 1919, at the end of the war, to work again with his old master at the Cavendish Laboratory where in 1923 became research vice-director. At the outbreak of war in 1914, as an English citizen, he had been interned in the Zivilgefangenenlager of Ruhleben. When Chadwick read the paper of the Joliot Curie couple, he went to speak with Rutherford who immediately answered "I do not believe it!" As a matter of fact, in order to expel high-speed protons from paraffin, the γ rays should have had energies of the order of 50 million eV. At this point, Chadwick repeated the Bothe–Becker experiment, bombarding the beryllium with α particles, and succeeded in proving that the emitted rays were made of particles. By measuring (indirectly) their mass, he showed that the mysterious rays, emitted according to the nuclear reaction

$$
{}_{2}^{4}\text{He} + {}_{4}^{9}\text{Be} \rightarrow {}_{6}^{12}\text{C} + {}_{0}^{1}\text{n}
$$

were not γ rays but the neutrons predicted by Rutherford.

Soon afterward Chadwick also discovered how to obtain neutrons by bombarding boron with α particles:

$$
{}_{2}^{4}\text{He} + {}_{5}^{11}\text{B} \rightarrow {}_{7}^{14}\text{N} + {}_{0}^{1}\text{n}
$$

The discovery of the neutrons, sent on 27 February 1932, to Nature and successively to other scientific journals (Chadwick [1932](#page-28-0)) procured for Chadwick the 1935 Nobel Prize for physics and offered the nuclear physicists a kind of projectile much more convenient than α particles or protons, since, being neutral, they were not repelled by the nuclear charge. After the appearance of neutrons on the physics scene, it became common practice to utilize the mass number, i.e., the sum of the number of protons and neutrons in place of the atomic mass of the isotopes.

6.6 Transuranium Elements

When the periodic table had been completed from hydrogen to uranium on the basis of Moseley's law, the problem arose of the existence of elements of atomic weight greater than that of uranium, i.e., of transuranium elements. Once it was understood that unstable elements existed, able to spit pieces off their nucleus in the form of α or β particles, releasing great amounts of energy as γ rays, it was natural to pose the question whether it was possible to profit from this instability to create new elements, bombarding atoms with projectiles able to smash their structure or even possibly to be swallowed in their nucleus, thereby increasing the atomic weight. At Rome in 1934 the Italian physicist Enrico Fermi (1901–1954) decided to profit from the fact that the neutrons had no electrical charge and to bombard uranium atoms, hoping to obtain new transuranium elements. Fermi and coworkers found that at least four new radioactive substances were formed (Fermi et al. [1934;](#page-29-0) Amaldi et al. [1935\)](#page-28-0) and assumed, perhaps too hastily, that among them new transuranic compounds could be present. Fermi's idea that bombarding uranium with neutrons could produce transuranium elements was soon criticized by Ida Noddack. She insisted that it was easier for a nucleus filled with protons and neutrons to split into smaller pieces rather than to absorb new neutrons, namely, that the fission of the nucleus was more probable than the formation of transuranic elements. Noddack's objection was, however, completely ignored by the physicists, not only because of the scientific prestige of Fermi but also because of the doubts arising from her false discovery of masurium.

As we shall see, both Fermi and Noddack were actually right. In 1939, Otto Hahn, Lise Meitner, and Fritz Strassmann realized at Berlin the fission of uranium, bombarded with neutrons. Later it was understood that the fission was the consequence of the formation of an unstable transuranic element that decayed into smaller fragments.

Fermi's idea was soon developed by Otto Hahn (1879–1968) in Berlin and led to the development of nuclear fission from which came both the atomic pile and the atomic bomb.

Otto Hahn, son of a rich, well-to-do glazier of Frankfurt am Main, studied chemistry at Munich and Marburg where in 1901 he received his Ph.D. in organic chemistry. He was not destined to study radioactive elements, being bent on a career as an industrial chemist. After the Ph.D., he went to England to improve his English and to visit Ramsay's laboratory in London where he found himself fully immersed in the world of nuclear physics and in direct contact with the principal representatives of this new branch of science.

Ramsay, who had at his disposal an impure sample of radium, asked him to purify it, and during this work Hahn discovered a new radioactive substance that he named radium-thorium. Excited by the discovery and encouraged by Ramsay, Hahn decided to forget his plans to join the chemical industry and decided to continue instead with the study of radioactive substances. To improve his competencies in this new field, he went to work from the autumn of 1905 to the summer of the following year with Rutherford at the McGill University in Canada. In this very profitable year, he discovered radioactinium, and learned how to study the emission of α particles from radioactive compounds, publishing a paper on their mass (Hahn 1908). Back in Germany he obtained a position at the Emil Fischer's Institute in Berlin, where he started his academic career. At the end of 1907, he met an Austrian theoretical physicist, Lise Meitner, Ph.D., from the University of Vienna, who had

visited Berlin to follow Planck's lectures. She started a collaboration with him that lasted more than 30 years. For some years they worked together in temporary rooms, since the university did not accept that a woman could have an official position, until in 1912 they moved to the new Institute of Chemistry of Kaiser Wilhelm Gesellschaft (today Max Planck Gesellschaft) at Berlin-Dahlem where Fritz Haber was the director of the Chemical Physics Institute and where Hahn became director of the Institute of Radiochemistry. At the outbreak of war, Hahn was recalled to arms and sent, together with James Franck and Gustav Hertz, to a special unit of the German army directed by Fritz Haber, specializing in the production of chlorine and mustard gases, while Lise Meitner became a Red Cross nurse, in charge of the x-ray service of the Austrian army.

At the end of the war they restarted their collaboration in Berlin and in 1918 discovered protactinium 231, the mother element of the actinides series (Hahn and Meitner [1918\)](#page-29-0). In 1921, Hahn discovered uranium Z, the first example of nuclear isomerism, i.e., the first example of the existence of a metastable nucleus theoretically explained in 1936 by the physicist and philosopher Carl Friedrich von Weizsäcker (1912–2007), a former assistant of Lise Meitner who was later involved in the uranium project for the construction of a German atomic bomb.

For more than 12 years Hahn dedicated himself to the study of the application of radioactive techniques to chemical problems until, at the beginning of 1938, with Lise Meitner and his assistant Fritz Strassmann (1902–1980), he decided to continue the kind of research started by Fermi in Italy, bombarding uranium with neutrons. Unfortunately with the annexation of Austria to Nazi Germany, Lise Meitner, of Jewish origin, was forced to leave Germany, and with the help of Niels Bohr was able to immigrate clandestinely to Sweden where she met her nephew, Otto Frisch, also a theoretical physicist. Before leaving Berlin, she published her last paper in collaboration with Hahn and Strassmann, maintaining that she had produced a new transuranic element with a half-life of 60 days, probably an isotope of iridium (Meitner et al. [1937\)](#page-30-0).

After Meitner's departure, Hahn and Strassmann continued the experiments of bombardment with neutrons while still keeping in touch with Lise, the only true theoretician of the group. Lise Meitner and her nephew Otto Frisch understood Hahn's problem and wrote a letter to him suggesting that he should determine whether barium had been formed as a consequence of the bombardment of uranium with neutrons, since in this case the uranium had been broken into pieces. The research on barium, made using an organic salt of barium obtained from the chemist Wilhelm Traube (1866–1942), gave positive results and thus, at the end of 1938, Hahn and Strassmann communicated that as consequence of bombardment with neutrons the uranium atom had broken into two pieces (Hahn and Strassmann [1938](#page-29-0)). In further papers published in 1939 they announced that the other product of the fission was krypton (Hahn and Strassmann [1939a](#page-29-0)) and also that they had realized the fission of thorium (Hahn and Strassmann [1939b\)](#page-29-0). At the same time, the theoretical interpretation of the nuclear fission process developed in Sweden by Lise Meitner and her nephew Otto Frisch appeared in Nature (Meitner and Frisch [1939\)](#page-30-0).

For the discovery of nuclear fission, Hahn obtained the 1944 Nobel Prize in chemistry, a prize that he could collect only in 1946 when he went back to Germany, since at the end of the war he had been interned at Farm Hall, Godmanchester, close to Cambridge in England, being suspected to having contributed to the development of the German atomic bomb.

After the demonstration that the bombardment of the nucleus with neutrons gave rise to fission, as predicted by Noddack, the proof also arrived that transuranic elements could be formed as maintained by Fermi. In 1931, at Berkeley in California, Ernest Orlando Lawrence (1901–1958) constructed the first cyclotron (Lawrence and Livingston [1931](#page-30-0)), able to accelerate protons at energies of one million eV and in 1932, with the creation of the Radiation Laboratory, he succeeded in producing protons with energies of about four million V. With this new instrument, Lawrence discovered the transmutation of sodium through bombardment with deuterium atoms. In 1940, Edwin Mattison McMillan (1907–1991), in collaboration with Philip H. Abelson, discovered the first transuranic element, neptunium 239, bombarding uranium 238 with slow neutrons produced by the Berkeley cyclotron (McMillan and Abelson [1940\)](#page-30-0):

$$
{}^{238}_{92}\text{U} + {}^{1}_{0}\text{n} \longrightarrow {}^{239}_{92}\text{U} \longrightarrow {}^{239}_{93}\text{Np}
$$

and in 1940 Glenn T. Seaborg, Edwin M. McMillan, J.W. Kennedy, and A.C. Wahl discovered that neptunium 239 has a half-life of 2 days, one half decaying into plutonium by the emission of β electrons:

$$
^{239}_{93}Np \longrightarrow ^{ \beta}_{94}Pu
$$

The discovery of plutonium was kept secret up to the publication of the paper at the end of the war (Seaborg et al. [1946\)](#page-31-0).

The discovery of plutonium, for which McMillan and Seaborg received the 1951 Nobel Prize for physics, went far beyond their imagination and expectations. It was a discovery that has changed the course of history with the creation of the most terrible instrument of death that the human mind has devised – the atomic bomb.

In 1942, Enrico Fermi and his group at Chicago transformed uranium 238 into plutonium through a chain reaction that they achieved inside the first nuclear reactor. With the development in 1944 of the Manhattan project for the construction of the atomic bomb, research on nuclear fission increased very rapidly. Among the chemists involved in the Manhattan project, two researchers, Glen Seaborg and Albert Ghiorso, assumed a leading position in the research of transuranic elements and succeeded in giving back to inorganic chemistry the fundamental role that it had in the past.

Glen Seaborg (1912–1999) started his academic career at Berkeley under the supervision of Gilbert Newton Lewis with a Ph.D. thesis on the inelastic scattering of fast neutrons and afterward worked in inorganic chemistry and in particular in the search for the isotopes of the elements and for new transuranic elements.

He followed closely the developments that Enrico Fermi's group realized in Italy by bombarding uranium with neutrons, and the research that Otto Hahn performed in Berlin on the transuranic elements.

At Berkeley, he performed important research on artificial radioactivity, collaborating with the physicist John J. Livingood to use the newly completed Lawrence Radiation Laboratory 37-inch cyclotron to produce and discover several dozen new isotopes. During his lifetime he identified more than 100 isotopes of different elements. In 1937, in collaboration with John Livingood and Fred Fairbrother, he created isotope 59 of iron (^{59}Fe) , later widely utilized in research on hemoglobin and in 1938 the isotope 131 of iodine (^{131}I) , a very important nucleotide in the cure of thyroid diseases (Livingood and Seaborg [1938](#page-30-0)).

Together with his coworkers at the Lawrence Laboratory that he directed, and in particular with the electro-technical engineer Albert Ghiorso (1915–2010) who later succeeded him as director of the group, he prepared as many as ten transuranic elements (Ghiorso et al. [1950](#page-29-0)).

In 1940, Seaborg prepared plutonium 239 (^{239}Pu) by bombardment of uranium with deuterons. The result was officially published only in 1946 (Seaborg et al. [1946\)](#page-31-0) due to the limitations on publication of material of military interest during the war. In 1944, he isolated americium 241 (^{241}Am) , bombarding plutonium 239 with α particles in a nuclear reactor, obtaining in succession the isotopes ²⁴⁰Pu and ²⁴¹Pu. Plutonium 241 transformed into 241 Am by β decay. In 1944, Seaborg prepared curium (242 Cm) by bombarding plutonium with α particles according to the nuclear reaction

$$
^{239}\text{Pu} + ^4\text{He} \rightarrow ^{242}\text{Cm} + ^1\text{n}.
$$

In 1949, he isolated berkelium (243 Bk) by bombarding americium with α particles and producing 241 Am plus two neutrons. In 1950, he synthesized californium (245 Cf) by bombarding curium (242 Cm) with α particles and producing 245 Cf plus one neutron (Seaborg et al. [1950\)](#page-31-0). In 1952, he identified fermium (255 Fm) and einsteinium (^{253}Es) in the debris of the explosion of the first atomic bomb and accounted for a complex nuclear reaction in which 15 neutrons were absorbed by uranium 238, giving rise to a chain of 7 beta decays (Ghiorso et al. [1955b](#page-29-0)). In 1955, Berkeley's group produced mendelevium $(^{256}$ Md) plus one neutron, bombarding a sample of einsteinium with α particles (Ghiorso et al. [1955a](#page-29-0)). In 1958, with the aid of a linear accelerator Berkeley bombarded curium with carbon ions, obtaining the isotope 254 of element 102 named nobelium $(^{102}$ No) (Ghiorso et al. [1958](#page-29-0)). Finally, in 1974, Berkeley's group and independently a group of soviet researchers of the Joint Institute for Nuclear Research at Dubno, directed by the physicist Georgij Nikolaevič Flerov (1913–1990), discovered the isotopes of mass 263 and 259 , respectively, of element 106 (Ghiorso et al. [1974;](#page-29-0) Oganesian et al. [1974](#page-30-0)).

An important theoretical contribution of Seaborg was the identification of the series of the actinides separated from the rest of the periodic table as that of the lanthanides. Seaborg reached this conclusion after a series of vain attempts to synthesize americium and curium.

The synthesis of mendelevium, nobelium, fermium, and einsteinium gave rise to long nationalistic disputes between the United States and the Soviet Union where the group of nuclear physicists working at Dubno had obtained the same results and boasted priority.

After the Second World War, Seaborg became one of the most prominent American physicists, deeply involved in the nuclear energy policy and in the production of nuclear weapons as adviser of several presidents from Truman to Clinton. From 1961 to 1971 he was president of the United States Atomic Energy Commission.

References

- Adloff J-P, Kauffman GB (2005) Francium (Atomic Number 87), the last discovered natural element. Chem Educ 10(5):387–394
- Ambarzumian V, Iwanenko D (1930a) Les électrons inobservables et les rayons. Compt Rend Acad Sci Paris 190:582
- Ambarzumian V, Iwanenko D (1930b) Über eine Folgerung der Diracschen Theorie der Protonen und Elektronen. Dokl Akad Nauk SSSR-A 6:153–155
- Amaldi E, d'Agostino O, Fermi E, Pontecorvo B, Rasetti F, Segre E (1935) Artificial radioactivity produced by neutron bombardment. II. Proc R Soc A 149:522
- Aston FW (1919) Isotopes and atomic weights. Nature 104:393
- Aston FW (1920a) The constitution of the elements. Nature 105:617
- Aston FW (1920b) The mass spectra of chemical elements. Philos Mag 39:611
- Aston FW (1922) Isotopes. Edward Arnold & Co., London
- Aston FW, Baxter GP, Brauner B, Debierne A, Leduc AF, Richards TW, Soddy F, Urbain G (1923) Report of the international committee on chemical elements. J Am Chem Soc 45:867
- Aston FW (1937) A second-order focusing mass spectrograph and isotopic weights by the doublet method. Proc R Soc Lond A 163:391–404
- Becquerel H (1896a) Sur les radiations emises par phosphorescence. Compt Rend 122:420–421
- Becquerel H (1896b) Sur les radiations invisibles emises par les corps phosphorescents. Compt Rend 122:501–503
- Becquerel H (1901) Sur la radioactivité de l'uranium. CR Acad Sci 83:977–978
- Brauner B (1901) Die basis der Atomgewichte. (IV. Abhandlung.). Z Anorg Chem 26:186–202
- Brauner B (1926) The new element of atomic number 61: illinium. Nature 118:84–85
- Chadwick J (1932) Possible existence of a neutron. Nature 129:312, The existence of a neutron. Proc R Soc Ser A 136:692
- Curie E (1938) Madame Curie. Gallimard, Paris
- Curie J, Curie P (1880) Développement, par pression, de l'électricité polaire dans les cristaux hémièdres à faces inclinées. CR Acad Sci 91:294–295
- Curie M (1899) Sur le poids atomique du métal dans le chlorure de baryum radifère. Compt Rend 129:760–762
- Curie P (1895) Lois expérimentales du magnétisme. Propriétés magnétiques des corps à diverses temperatures. Ann Chim Phys 5:289–405
- Curie P, Curie M (1898) Sur une nouvelle substance radioactive, contenue dans la pechblende. CR Acad Sci 127:175–178
- Curie P, Curie M, Be`mont G (1898) Sur une nouvelle substance fortement radioactive contenue dans la pechblende. CR Acad Sci 127:1215–1217
- Curie P, Curie M (1899) Sur la radioactivite provoquee par les rayons de Becquerel. CR Hebd Acad Sci 129:714–716
- Curie I, Joliot F (1932) Émission de protons de grande vitesse par les substances hydrogénées sous
- l'influence des rayons γ trés pénétrants. Compt Rend 194:273
Debierne AL (1899) Sur une nouvelle matiére radio-active. Compt Rend 129:593
- Debierne AL (1903) Sur la radioactivité induite provoquée par les sels d'actinium. Compt Rend 136:446
- Debierne AL (1904) Sur l'actinium. Compt Rend 139:538–540
- De Gregorio A (2006) Enrico Fermi and the discovery of neutron-induced radioactivity: a project being crowned. Stud Hist Philos Mod Phys 37:330
- Dorn FE (1900) Die von radioactiven Substanzen ausgesandte Emanation. Abhandlungen Naturforschenden Gesellschaft (Halle) 23:1–15
- Fajans K (1913a) Die radioaktiven Umwandlungen und das periodische System der Elemente. Berichte 46:422–439
- Fajans K (1913b) A relationship between the nature of a radioactive transformation and the electrochemical behavior of the respective elements. Phys Z 14:131
- Fermi E, Amaldi F, d'Agostino O, Rasetti F, Segre` E (1934) Artificial radioactivity produced by neutron bombardment. I. Proc R Soc A 146:483
- Ghiorso A, James RA, Morgan LO, Seaborg GT (1950) Preparation of transplutonium isotopes by neutron irradiation. Phys Rev 78:472
- Ghiorso A, Harvey B, Choppin G, Thompson S, Seaborg G (1955a) New element mendelevium, atomic number 101. Phys Rev 98:1518
- Ghiorso A, Thompson SG, Higgins GH, Seaborg GT, Studier MH, Fields PR, Fried SM, Diamond H, Mech JF, Pyle GL, Huizenga JR, Hirsch A, Manning WM, Browne CI, Smith HL, Spence RW (1955b) New elements einsteinium and fermium, atomic numbers 99 and 100. Phys Rev 99:1048–1049
- Ghiorso A, Sikkeland T, Walton JR, Seaborg GT (1958) Element 102. Phys Rev Lett 1:17–18
- Ghiorso A, Nitschke JM, Alfonso JR, Alonso CT, Nurmia M, Seaborg GT, Hulet EK, Lougheed RW (1974) Element 106. Phys Rev Lett 33:1490–1493
- Giauque WF, MacDougall DP (1933) Attainment of temperatures below 1° absolute by demagnetization of $Gd_2(SO_4)$ ₃·8H₂O. Phys. Rev. 43:768
- Giauque WF, MacDougall DP (1935) The Thermodynamic Temperature Scale in the Region Below 1° Absolute, Phys. Rev. 47, 885–886
- Giauque WF, Johnston HL (1929a) An isotope of oxygen of mass 17 in the Earth's atmosphere. Nature 123:831
- Giauque WF, Johnston HL (1929b) An isotope of oxygen, mass 18. J Am Chem Soc 51:1436–1441
- Giesel FO (1900) Über radioactive Stoffe. Ber Deut Chem Ges 33:3569
- Giesel FO (1902) Über radium und radioactive Stoffe. Ber Deut Chem Ges 35:3608–3611
- Giesel FO (1903) Über den Emanationskörper aus Pechblende und über Radium. Chem Ber 36:342
- Giesel FO (1904) Über emanium. Ber Deut Chem Ges 37:1696–1699
- Giesel FO (1905) Über Emanium. Ber Deut Chem Ges 38:775–778
- von Grosse A (1934) Element 91. Science 80:512–516
- Hahn O (1908) Ein kurzlebiges Zwischenprodukt zwischen Mesothor und Radiothor. Phys Z 9:246
- Hahn O, Meitner L (1918) Die Muttersubstanz des Actiniums, ein Neues Radioaktives Element von Langer Lebensdauer. Phys Z 19:208
- Hahn O (1936) Applied radiochemistry. Cornell University Press, Ithaca
- Hahn O, Strassmann F (1938) Die chemische Abscheidung der bei der Spaltung des Uran. Naturwissenschaften 26:756
- Hahn O, Strassmann F (1939a) Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans mittels Neutronen entstehenden Erdalkalimetalle. Naturwissenschaften 27:11
- Hahn O, Strassmann F (1939b) Nachweis der Entstehung activer Bariumisotope aus Uran und Thorium durch Neutronenbestrahlung; Nachweis weiterer aktiver Bruchtucke bei der Uranspaltung. Naturwissenschaften 27:89–95
- Hopkins BS, Yntema L, Harris JA (1926) The element of atomic number 61; illinium. Nature 117:792
- Iwanenko DD (1932) The neutron hypothesis. Nature 129:798
- Lawrence EO, Livingston MS (1931) The production of high speed protons without the use of high voltages. Phys Rev 38:834
- Livingood JJ, Seaborg GT (1938) Radioactive iodine isotopes. Phys Rev 53:1015
- Mayer H (1904) Die neueren Strahlungen; Kathoden-, Kanal-, Roentgen-Strahlen und die radioaktive Selbststrahlung (Becquerelstrahlen), 1st edn. Mährisch-Ostrau R. Papauschek
- McMillan EM, Abelson PH (1940) Radioactive element 93. Phys Rev 57:1185
- Meitner L, Frisch OR (1939) Disintegration of uranium by neutrons: a new type of nuclear reaction. Nature 143:239–240
- Meitner L, Hahn O, Strassmann F (1937) Uber die umwandlungs Reihen des Urans die durch Neutronen Bestrahlung erzeugt verden. Z Phys 106:249
- Meyer, Stefan and Schweidler, Egon von (1904) "Influence of Alternations of Temperature on Radioactive Substances." Pysicalische Zeitschrift, 5:319–320
- Meyer S, von Schweidler E (1905) Sitzungberichte der Akademie der Wissenschaften zu Wien, Mathematisch-Naturwissenschaftliche Classe, p 1202 (Table 5)
- Moseley HGJ (1913) The high frequency spectra of the elements, part 1. Philos Mag 26:1024
- Moseley HGJ (1914) The high frequency spectra of the elements, part II. Philos Mag 27:703–713 Noddack W, Tacke I, Berg O (1925) Die Ekamangane. Naturwissenshaften 13:567
- Noddack W, Noddack I (1927) Über den Nachweis der Ekamangane. Z Angew Chem 40:250
- Oganesian YT, Tret'yakov YP, Il'inov AS, Demin AG, Pleve AA, Tret'yakova SP, Plotko VM, Ivanov MP, Danilov NA, Korotkin YS, Flerov GN (1974) Synthesis of neutron deficient isotopes of fermium,kurchatovium, and element 106. ZhETF Pis Red 20:580–585
- Owens RB (1899) Thorium radiation. Philos Mag 48:360
- Pais A (1991) Niels Bohr's times: in physics, philosophy, and polity. Oxford University Press, Oxford
- Paneth FA (1962) The epistemological status of the chemical concept of element. Br J Philos Sci 13.1
- Perey M (1939) Sur un élément 87, dérivé de l'actinium. Compt Rend 208:97
- Perrier C, Segrè E (1937) Alcune proprietà chimiche dell'elemento 43. Rend Lincei 25:723, serie 6
- Quinn S (1995) Marie Curie a life. Heinemann, London
- Ramsay W, Soddy F (1903) Experiments in radioactivity, and the production of helium from radium. Proc R Soc 72:204–207
- Ramsay W, Whytlaw-Gray RW (1910) Density of the emanation of radium. Compt Rend 151:126
- Richards TW, Lembert ME (1914) The atomic weight of lead of radioactive origin. J Am Chem Soc 36:1329–1344
- Rolla L, Fernandes L (1926) Über das Element der Atomnummer. Z Anorg Allg Chem 157:371
- Rutherford E (1899) Uranium radiation and the electrical conduction produced by it. Philos Mag 47:109–163
- Rutherford E (1900a) A radioactive substance emitted from thorium compounds. Philos Mag 49:1–14
- Rutherford E (1900b) Radioactivity produced in substances by the action of thorium compounds. Philos Mag 49:161
- Hahn O (1907) Mass of the α -particles from thorium. Philos Mag 12:371–378
- Rutherford E, Soddy F (1902a) The radioactivity of thorium compounds I. Trans Chem Soc 81:321–350
- Rutherford E II (1902) The radioactivity of thorium compounds II. The cause and nature of radioactivity. Trans Chem Soc 81:837–860
- Rutherford E, Soddy F (1902b) On the cause and nature of radioactivity. PhilosMag 4 (19–24):370–396
- Rutherford E, Geiger H (1908) The charge and nature of the alpha particle. Proc R Soc A81:162
- Rutherford E, Royds T (1909) The nature of the α particle from radioactive substances. Philos Mag 17:281
- Rutherford E (1920) Nuclear constitution of atoms. Proc R Soc Ser A97:374
- Rydberg JR (1886) Die Gesetze der Atomgewichtzahlen. Svenska Vetenkaps Academiens Handlungar 11:13
- Schmidt GC (1898) Über die vom Thorium und den Thorverbindungen ausgehende Strahlung. Ann Phys 65:141
- Scerri ER (1994) Prediction of the nature of hafnium from chemistry, bohr's theory and quantum theory. Ann Sci 51:137
- Seaborg GT, McMillan EM, Kennedy JW, Wahl AC (1946) Radioactive element 94 from deuterons on uranium. Phys Rev 69:366–367
- Seaborg GT, Thompson SG, Street K Jr, Ghiorso A (1950) Element 98. Phys Rev 78:298–299
- Soddy F (1913) The radio-elements and the periodic law. Chem News 107:97–99
- Thomson JJ (1913a) On the appearance of helium and neon in vacuum tubes. Nature 90:645 Thomson JJ (1913b) Rays of positive electricity. Proc R Soc A 89:1–20
- Urey HC, Brickwedde FG, Murphy GM (1932a) A hydrogen isotope of mass 2. Phys Rev 39:164–165
- Urey HC, Brickwedde FG, Murphy GM (1932b) A hydrogen isotope of mass 2 and its concentration. Phys Rev 40:1–15
- van den Broek A (1911) The number of possible elements and Mendeléeff's "Cubic" periodic system. Nature 87:78
- van den Broek A (1913a) Die Radioelemente, das periodische System und die Konstituenten der Atome. Phys Z 14:32–41
- van den Broek A (1913b) Intra-atomic charge. Nature 92:372–373
- Villard P (1900) Sur la transparence de l'aluminium pour le rayonnement du radium. Compt Rend 130:1154–1157
- Wood RW (1904) The N-rays. Nature 70:530–531