

History of Chemistry

DOI: 10.1002/anie.201008063

# Marie Curie: Recipient of the 1911 Nobel Prize in Chemistry and Discoverer of the Chemical Elements Polonium and Radium

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chemical elements  $\cdot$  Curie, Marie  $\cdot$  history of chemistry  $\cdot$  Nobel prize  $\cdot$  radioactivity

"One has to have persistence, but above all else belief in oneself. One has to believe that one has the talent to reach a certain goal and one can reach that goal no matter what it costs."[1]

After Marie Curie received the Nobel Prize in Physics in 1903, along with Antoine Henri Becquerel (1852–1908) and her husband Pierre Curie (1859–1906),<sup>[3]</sup> she received the Nobel Prize in Chemistry in 1911 (Figure 1) "in recognition of



Figure 1. Marie Curie's 1911 Nobel Prize certificate.

her services to the advancement of chemistry by the discovery of the elements radium and polonium, the isolation of radium, and the study of the nature and compounds of this remarkable element". [2] Besides Linus Pauling (1901–1994), she is the only person to receive Noble Prizes in different fields and the only woman among the four multiple Nobel Prize winners. [4]

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### Radiation of Uranium and Its Consequences

In the last third of the 19th century, classical physics had reached such an extent of completion that many believed that it was a completed science. However, phenomena discovered in the 1890s such as X-rays, anode rays, and radiation from uranium could not be explained by the theories of that time. On March 1, 1896, the French physicist Antoine Henri Becquerel of the Paris Academy of Sciences reported the observation that uranium emits a highly penetrating, invisible radiation that had an ionizing character. He, however, could not explain where uranium got the energy to emit radiation of seemingly undiminishing intensity. Shortly thereafter, he devoted himself to other areas of scientific investigation.

The Polish physicist, Marie Curie, living in Paris at the time, chose research on the radiation from uranium as the topic for her doctorate despite Bequerel's abandoning the topic. She spearheaded the use of a specialized electrometer that her husband, physicist Pierre Curie, and his brother, Jacques Curie (1855-1941), had constructed. The electrometer was capable of measuring the low electric current that flows when irradiated air became conducting. The basis of its operation was the piezoelectric effect.<sup>[7]</sup> Marie Curie's systematic experiments on an extensive collection of chemicals and minerals from the Natural History Museum revealed that thorium also emits radiation comparable to that of uranium. Approximately contemporary with, but independently from Marie Curie, the Berlin chemist, Gerhard Carl Schmidt (1865-1948), demonstrated the "Radiation of Thorium".[8] For this novel property of uranium and thorium Marie introduced the term "radioactivity" (Fr. "radioactivité") in 1898.<sup>[9]</sup> Experiments by the British physicist Ernest Rutherford (1871-1937) showed that the radiation was not homogeneous; its three components were named  $\alpha$ -,  $\beta$ -, and γ-rays (Figure 2).<sup>[10]</sup>

In February 1898 Marie Curie observed something completely unexpected: exactly two uranium minerals, pitch-blende (uranium oxide) and torbinite (copper uranyl phosphate) showed appreciably higher radioactivity than pure uranium. After the exclusion of measurement errors and methodological shortcomings, there was only one explanation. The minerals must contain an unknown element that has



# Production of Radium Chloride from the Residue of a Uranium Extraction, Ref. [7], pp. 24–25

"The residue contains mainly sulfates of lead and calcium, also silicon, aluminum, and iron oxides are present. Additionally, larger or smaller amounts of almost all metals are found (copper, bismuth, zinc, cobalt, manganese, nickel, vanadium, antimony, thallium, and the rare earths, niobium, tantalum, arsenic, barium, etc.). Radium is found in this mixture of sulfates and is the least soluble. To dissolve it as much sulfuric acid as possible must be removed. The treatment of the residue begins with a concentrated, boiling sodium hydroxide solution. The sulfuric acid containing lead, aluminum, and calcium goes into solution mainly as sodium sulfate, which is removed by washing with water. Lead, silicon, and aluminum are simultaneously removed with alkali. The insoluble part is then washed with water and the effect of the usual hydrochloric acid is skipped. This operation effects complete clarification of the substance and by and large it is dissolved. Out of this solution one can separate polonium and actinium: the former precipitates upon treatment with hydrogen sulfide, and the latter is found in the hydrates that precipitate upon treatment with ammonia after these are separated from the sulfates and oxidized. Radium remains in the insoluble part. This part is washed with water, then treated with concentrated, boiling sodium hydroxide solution. Only when a few unreactive sulfates remain behind, has this operation facilitated a complete transformation of barium sulfate into its carbonate. Then, the substance is thoroughly washed with water and subjected to the effect of hydrochloric acid, which must be thoroughly free of sulfuric acid. The solution, which contains barium as well as polonium and actinium, is filtered and precipitated with sulfuric acid. In this way one obtains a crude sulfate of barium containing radium and also calcium, lead, iron, and a trace of actinium. The solution still contains a little actinium and polonium, which can be separated in the same way as from the first hydrochloric acid solution."

a much higher activity than uranium. The search proved to be very difficult and expensive. The unknown substance was named "polonium" in honor of Madame Curie's birthplace. It was obtained in ppm amounts in the final product, whereas the initial raw material was in tons. Measurement of the radiation intensity of the various fractions with the abovementioned electrometer was helpful in the chemical separation. Staring in March 1898 Pierre Curie also participated in this work (Figure 3).

The Curies were supported with chemicals by the chemist Gustave Bémont (1857–1937) who was active in the École de Physique et Chimie. The raw material could be obtained from

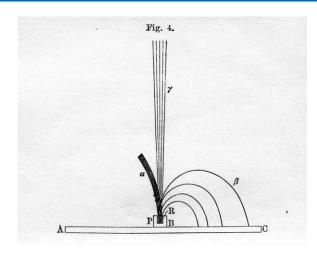


Figure 2.  $\alpha$ -,  $\beta$ -, and  $\gamma$ -rays.

the Bohemian town of St. Joachimstal (today Jáchymov). At that time it was part of the Austro-Hungarian empire and the only place in the world where uranium ore could be mined. The remaining residue after the extraction of uranium from pitchblende was forwarded to Paris by the Austro-Hungarian government free of charge. (Only the transportation costs had to be paid.). Uranium was used as a coloring agent to produce a yellow-green color in glass. Polonium was discovered in July 1898. [11] Soon after, it was evident that the raw material had a second emitter; it was found in December 1898 and given the name "radium". [12] Finally, in December 1899 the physicist André Louis Debiere (1874–1949) discovered still another new element which he named "actinium". With this, he confirmed a suspicion of Marie Curie that there was another radioactive element. [13]

In March 1902 the Curies had isolated approximately 100 mg of radium chloride. Marie determined the value of the atomic weight of radium to be  $225 \pm 1$  (today's value is 226.0254) and placed it in the Periodic Table in the group of alkaline-earth metals (under barium).[14] She used these results in her dissertation "Recherches sur les substances radioactives" (Investigations on Radioactive Substances). On June 25, 1902 Marie Curie became the first woman to graduate from the Parisian Sorbonne University (founded in the 12th century) with a Doctor's degree in the natural sciences. The result of her oral examination was "très honorable". In the following year the Curies together with Becquerel received the Nobel Prize in Physics. Originally only Bequerel and Pierre Curie were candidates as prize recipients, but the Swedish mathematician Magnus Goesta Mittag-Leffler (1846–1927) made sure that Marie was also awarded the prize.<sup>[15]</sup>

## Background and Education<sup>[16]</sup>

Marie Curie was born on November 7, 1867 as Maria Salomea Skłodowska in Warsaw. Her parents—she was the fifth and last child—were descendents of poverty-stricken Polish gentry. Her father, Władislaw Skłodowski, an assistant superintendent of an academic secondary school, was re-





Figure 3. Pierre and Marie Curie in the laboratory.

moved from his post in 1873 in Russian-occupied Warsaw on political grounds. [17] He was a passionate teacher and was especially interested in the natural sciences. From 1860 her mother was the principal of a private girls school and, although she gave birth to five children in seven years, she was actively employed. She became severely ill with tuberculosis and died in 1878. For Marie, then just ten years old, it was "the first serious sorrow and the first big despair" in her life. [18]

At first Marie attended the girls school, where her mother was principal, and then a private school, but in 1878 she transferred to a girls academic secondary school, where she passed her exit examination as the best in her class. Afterward, she and her sister, Bronia Skłodowska, gave private instruction. The two educated themselves at the so-called "Flying University" (Figure 4), at which 1000 female students obtained an academic education despite a ban by the Russian authorities.

Marie took a position as a governess in the countryside so that her sister could study medicine in Paris. After the completion of her sister's studies, they wanted to switch roles. In 1889 Marie returned to Warsaw (Figure 5), where for the first time she had the opportunity to carry out experiments in the laboratory of the Warsaw Industry and Agriculture



Figure 4. The "Flying University" in Warsaw.



Figure 5. Portrait of Marie Curie as a young woman.

Museum with her cousin Józef Boguski (1853–1933) and a high school student, Dmitrii Ivanovič Mendeleevs (1834–1907).

In 1891 Marie Curie could finally go to Paris to study. She was one of the 23 female students of the "Faculté des Sciences", who overwhelmingly came from abroad, since neither natural sciences nor Latin or Greek were taught in French girls schools.<sup>[19]</sup> She registered herself in physics. Her teachers included outstanding professors such as Gabriel Jonas Lippman (1845–1921),<sup>[20]</sup> who received the Nobel Prize in 1908, Joseph Boussinesq (1842-1921), the mathematicians Paul Appell (1855-1904) and Henri Poincaré (1854-1912), and the chemistry professor Emil Duclaux (1840-1904). She devoted herself with great enthusiasm to her studies and passed her final examination in 1893 as the best in her class. With the help of von Lippmann she received 600 Francs from the Society for the Fostering of National Industry for a project on the analysis of the magnetic properties of different types of steel. Even with this support at her disposal, she still made only slow progress.

#### A Momentous Meeting

A Polish friend introduced Marie to Pierre Curie, who she asked for help in the procurement of laboratory space. Pierre Curie was already a successful scientist at this time. His studies on crystallography led him and his older brother Jacques to the discovery of piezoelectricity, while his work on symmetry in the field of magnetism led to the establishment of the Curie Law. Eve Curie attested "What was at first a lighthearted conversation became an intense scientific dialogue between Pierre Curie and Marie Sklodowska. Marie, a little shy, respectfully asked questions and listened to Pierre's enthusiastic answers. He explained his plans, described the phenomenon of crystal formation, which was currently occupying him and whose laws he was investigating." [21] In any



case, Pierre was not a member of the scientific establishment and taught only at the "École Municipale de Physique et Chimie Industrielle" where he had temporary access to a laboratory.

Although Marie saw herself as a patriotic Pole who would return to her homeland, she married Pierre in 1895. They spent their honeymoon on a bicycle tour through Brittanny (Figure 6).<sup>[22]</sup> In 1897 their first daughter Irène was born.



Figure 6. Pierre and Marie Curie with their bicycles.

Marie continued her scientific work despite familial duties; a second daughter, Eve, was born in 1904. A workshop on the ground floor of Pierre's school served as a laboratory. It was here that she began the experimental work for her PhD thesis.

#### On the Way to the Second Nobel Prize

The 1903 Nobel Prize received tremendous publicity from the press and made the husband-and-wife research team well known outside of France. The prize money of 70000 Francs enabled the Curies to hire an assistant. In 1904 Pierre accepted a professorship in general physics, specially designed for him, at the Sorbonne. Although at first his own dedicated laboratory was not envisioned, Members of Parliament finally made funds available for that endeavor and Marie led the scientific work as its "chef des travaux." [23] The planned trip to Stockholm was only possible in early 1905 because of Pierre's sickness due to handling radiating materials. Not until June 6, 1905 did Pierre give his long overdue Nobel Prize address to the Royal Academy of Sciences in Stockholm on the investigation of radioactivity. [24]

In the following year Marie Curie experienced a terrible stroke of fate. On April 19, 1906 Pierre had an accident while crossing a street. The left back wheel of a carriage rolled directly over his head and crushed his skull. He was instantly dead, and an exceedingly happy marriage and collaboration ended. Slowly, the needs of her children and her work brought

Marie Curie back to everyday life. She assumed the work of Pierre as a university lecturer and director of the laboratory and, then in 1908, as a professor. In her diary, which was first accessible in 1990, she related "They offered me to be your successor, my Pierre: your lectures and your laboratory. I accepted. I don't know if it's right or wrong. You often told me you would like to see me give lectures at the Sorbonne. And I want to at least try to continue our work." [25] Marie was the first woman in the long history of the Sorbonne to give lectures. [26] She was held in high esteem; the number of assistants and employees in her laboratory grew from seven to 24.[27] Nevertheless, she did not succeed in becoming a member of the prestigious Académie des Sciences because the majority of the researchers refused to accept a woman in their midst. In 1910 she collected her results in a two-volume book, "Traité de Radioactivité" (Figure 7).[28]

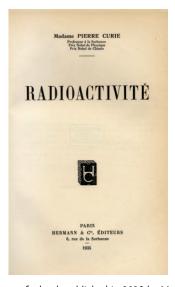


Figure 7. Title page of a book published in 1910 by Marie Curie.

In 1911 she was nominated for the Nobel Prize in Chemistry. At the 1911 Solvay Conference in Brussels—the Belgian industrialist Ernest Solvay (1838–1922) had developed a new process for the production of soda ash and invited the leading minds of the time to this conference—the guests included Max Planck (1856–1947), Ernest Rutherford (1871–1937), and Marie Curie. She received a telegram from the Nobel Prize Committee. In the letter from the committee, it was pointed out that she had been able "to produce a sample of radium so pure that its atomic weight could be determined and she had succeeded in 1910 to extract radium in its metallic state." [30] [Other researchers would subsequently confirm her value<sup>[29]</sup> (added by the authors)].

The award of the Nobel Prize in 1911 was overshadowed by the outing of her love affair with Paul Langevin (1872–1946). Langevin, student of her husband, five years younger than Marie and unhappily married, was a leading physicist and mathematician. He supported Marie in the preparation of her lectures and improved her presentation style. The French press led a real mudslinging campaign against her. After the



release of her diaries in the 1990s, the American Susan Quinn reworked this long-taboo chapter in the life of Madame Curie in her Curie biography. [31] Marie's extraordinary workload, her maternal responsibilities, and the media hype took its toll on her health. Despite her ill health she participated in the Nobel Prize Award Ceremony on December 10 and 11, 1911 in Stockholm. She gave a lecture in which she not only confidently presented her accomplishments but also pointed out the fundamental contributions of her deceased husband, and those of Ernest Rutherford (1871–1937) and Frederick Soddy (1877–1956). Finally, she summed up: "With this, there is a completely new kind of chemistry whose preeminent tool is the electrometer and not the balance. One will eventually name it the chemistry of the unweighable." [32]

After her return to Paris she was completely exhausted and very depressed. She was admitted to a hospital for a one-month stay, and a longer convalescence followed. Additionally, in March 1912 she had to undergo surgery. Slowly, Marie Curie recovered and eventually took on new tasks (Figure 8).



Figure 8. Marie Curie in the laboratory (Musée Curie in Paris) (CNRS/Institute Curie) 11, rue Pierre et Marie Curie).

#### The Radium Institute of Paris

Radium had, as the English dramatist George Bernard Shaw (1856–1950) remarked in the introduction to his 1906 comedy, "The Doctor's Dilemma", turned the world upside down, since "it challenged our devoutness as much as the apparitions of Lourdes challenged the devoutness of the Catholics." [33] It was soon shown that radioactive emitters had enormous physiological effects. There was a real boom: radium was mixed in many teas, creams, and bath salts; even in hair tonic as a remedy against hair loss, in a pouch carried next to the testicles to combat impotence, and in toothpaste proclaimed to whiten teeth. Marie Curie herself noticed that radium tended to make her hands scaly, stiffen her fingertips, and sometimes make them hurt. [34] The dermatologist Henri-Alexandre Danlos (1844–1912), chief physician at the Saint-

Louis Hospital in Paris, had successfully tested radium chloride as a medicine. He was a pioneer in his successful treatment of lupus erythematodes and skin tubercolosis with the use of radium salts.<sup>[35]</sup> Consequently, Marie Curie occupied herself with the medical, biological, and commercial uses of radioactivity and tried to quantify the energy of radium.<sup>[36]</sup>

A Radium Institute was founded in Paris in 1914 to investigate not only the physics and chemistry of radioactive elements but also their possible medical applications. Under Marie's leadership, the institute finally reached its potential in early 1919 after the First World War. After 1920 the scientific work of the Radium Institute was effectively supported by the Curie Foundation of the banker Henri de Rothschild (1872–1946).<sup>[37]</sup>

During the First World War Marie not only invested part of her Nobel Prize money in war bonds, she also organized and ran a mobile X-ray station for wounded soldiers with her 17-year-old daughter, Irène. Furthermore, she trained approximately 150 X-ray technicians by the end of the war (Figure 9).

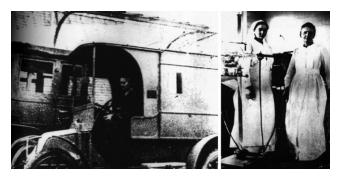


Figure 9. Left: X-ray vehicle. Right: Marie and Irène Curie in a mobile X-ray station in the First World War.

After the war, Marie Curie, in addition to her ongoing work at the Radium Institute, played an active part in the Institute of Intellectual Collaboration of the League of Nations. In May 1920 she visited the USA, where journalist Marie Meloney (1873–1943) took the opportunity to support her financially. She was received by the President of the United States and celebrated in the press. Meloney attended to spreading many a legend about Marie Curie and financed the purchase of 1 g of radium. [38] In the following period Marie Curie received many awards and honors among which were honorary doctorates from Edinburgh (1907), Geneva (1909), and Birmingham (1913). [39] In the year 1932 she was also selected to be a member of the present-day German Academy of Sciences Leopoldina—National Academy of Sciences (Figure 10). [40]

In her last years Marie would live to see that her daughter Irène (1897–1958) and her husband Fréderic Joliot (1900–1958) would continue the successful work at the Institute. They both received the Nobel Prize in 1935. In 1932 Marie Curie, severely sick, passed on the directorship of the Institute to her daughter Irène. On July 3, 1934 she fell into a coma and





*Figure 10.* Marie's Curie's letter to the President of the German Academy Leopoldina.

died one day later. The cause of death was given as "malignant anemia in extreme progress", the consequences of permanently high levels of radiation. Marie Curie worked with radium without protection and even pipetted solutions of radium and polonium with her mouth. She kept a test tube of radium salts next to her bed in order to have before her eyes the "beautiful glimmer" of her "child" as she fell asleep.<sup>[41]</sup>

#### Summary

Marie Curie's life was probably significantly influenced by her family. She understood how to persevere as a woman in the male-dominated scientific world, and she had seen how her mother had worked and met with success. As the daughter of a staunchly patriotic Polish father she became a French patriot during the First World War. Like her parents, she passionately devoted herself to her profession and deferred all else, her health, and also her family, to her research. She grew up in poverty, and her personal needs were exceedingly modest.

The determination and steadfastness with which Marie Curie pursued her scientific undertakings—the discovery of the elements, radium and polonium, as well as their isolation, investigation, and characterization, for which she won the Nobel Prize in 1911—can be an inspiration for young scientists today. Just as her persistence served as a model, the results of her research, in which she endeavored to explore the medical, biological, and commercial uses of radioactivity, served the well-being of her fellow humans. Her medical work during the First World War and her commitment to the Institute of Intellectual Cooperation of the League of Nations showed that she was not a remote researcher, but a politically engaged scientist. Marie Curie and her research had an extraordinary, definitive effect on the

chemistry of the 20th century, and her extreme idealism continues to inspire scientists today.

Received: December 21, 2010 Published online: March 29, 2011

Translated by Gordon Stevens, Berkeley, CA (USA)

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