"Chance only favors the prepared mind"—Pasteur

Becquerel's Discovery of Radioactivity in 1896

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The basic discovery, from which stemmed Nuclear Physics, Nuclear Chemistry, and Nuclear Medicine, occurred in Paris just 80 years ago.

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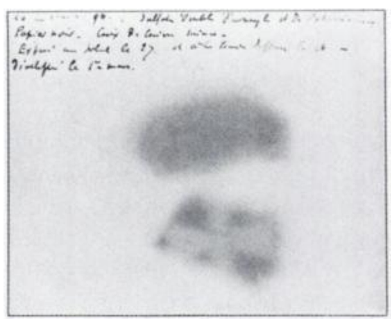
HENRI BECQUEREL (1852–1908)
(Photo. Bibliothèque Nationale, Paris)



Henri Becquerel's recognition of the significance of this first autoradiograph constituted his discovery of radioactivity in Paris on Sunday, 1 March 1896 (*1,2,4,5*). The image resulted from exposure of a photographic plate to radiations emitted spontaneously from crystals of a uranium salt. Today we know these chiefly were β-particles. Some were absorbed by crossed thin strips of copper he placed between the crystals and plate. Becquerel published the illustration in his Nobel Lecture in 1903 (*2*).

Becquerel sent this photograph in 1903 to the Nobel Foundation for publication in *Les Prix Nobel*.

On Sunday, 1 March 1896, Henri Becquerel developed photographic plates upon which he had placed thin transparent crystalline crusts of the double sulfate of uranium and potassium a few days previously. To his surprise, blackening of the emulsion had occurred beneath the positions of the crystals and he saw silhouetted images of the shapes of the crusts. *Within hours*, he had performed two well-designed confirmatory experiments which demonstrated clearly that radiations emitted from the crystals not only passed through the thick black paper enclosing the plates, but that they penetrated thin strips of aluminum and copper also. And even patterns were seen of metal screens interposed between the crystals and the plates (*1–5*).

Becquerel presented his discovery of radioactivity* *the following evening* at the next weekly meeting, held each Monday in Paris, of The French Academy of Sciences. His paper promptly was published in *Comptes rendus* (*1*) *within 10 days* (*5,6*).

In his next paper, only a week later, Becquerel already had eliminated substances other than those

* The apt term, *radioactivity*, was coined by Marie Curie. It first appeared in print (*3,7*) in the title of a paper she published in 1898 with her husband, Pierre.

containing uranium as the source of the radiation. He disclosed also that the penetrating rays, like x-rays, dissipated an electric charge on a gold-leaf electroscope (*5*). This gave him a second method to pursue his studies, one which provided numerical data, and he no longer was limited to the qualitative photographic method (*2*).

The paper resulting from his presentation at the séance on 23 March revealed his diligent explorations into the nature of the phenomenon he had discovered only 3 weeks previously. Intercomparisons by means of the electroscope of his rays with the penetrating radiation emitted from a Crookes tube (i.e., Röntgen rays) led him to hypothesize that they had different wavelengths. Uranium salts kept in darkness 15 days continued to emit the rays with essentially undiminished intensity. Water, and solutions of several substances, were relatively transparent, as were sulfur and paraffin, whereas thick aluminum and tin were penetrated much less readily. Uranyl nitrate, which was recrystallized after having been melted in its own water of crystallization in darkness throughout, exhibited no diminution in its photographic effect (*5*).

Becquerel demonstrated in his fifth presentation, on 18 May (*5,8*), that metallic uranium not only gave a severalfold greater effect photographically than did uranium salts, but that it more rapidly discharged the gold-leaf electroscope as well (*2,5*). Thus, his demonstration that *uranium* itself *specifically* was the source of the radiations completed his discovery (*5,8*). This was less than 3 months after his disclosure of their existence to The Academy on 2 March 1896.

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Like the discovery of Röntgen rays, Becquerel's discovery of the rays named after him (*2,9,10*) was a prime example of serendipity (*11*). The sagacious recognition by his "prepared mind" that something new and unexpected was being observed occurred during the course of an experiment designed to test another hypothesis, which thereby was proved untenable.

Becquerel had learned at the meeting of The Academy on 20 January 1896 that the propagation of penetrating x-rays, which first had been described by Professor Röntgen only 3 weeks previously, seemed to be associated with the intense fluorescence induced in the glass wall of a Crookes tube, at the spot where the cathode rays (electrons) struck it (*5*).

At the time of his discovery, (Antoine-) Henri Becquerel (*2,9,12*) was Professor of Physics at The Museum of Natural History in Paris, the same position held before him by his father Alexandre-Edmond and his grandfather Antoine-César. He was Professor also at the École Polytechnique and at the Conservatoire National des Arts et Métiers. Thus, he simultaneously held three chairs of physics in Paris (*12*). Among his endeavors was a continuation of the studies of phosphorescence begun when he served as an assistant to his father, who had investigated the phenomenon in uranium compounds (*9,11*).

When he learned of Röntgen's findings, Becquerel postulated that the penetrating x-rays might be interrelated with some transformation of the visible rays involved in phosphorescence. To test his hypothesis, he placed the crystalline crusts of potassium uranyl sulfate, inherited from his father (*2*), upon photographic plates enclosed in a double thickness of black paper* before exciting phosphorescence in the crystals in bright sunlight. Indeed, images of the crusts were found when he developed the plates, thus *apparently* supporting his hypothesis.

To continue these studies, he prepared additional plates and similarly placed on them the crusts of $K_2(UO_2)(SO_4)_2 \cdot 2H_2O$. Because the sun shone only intermittently, he put the assembly of the plates with the crystals in place upon them in a dark cabinet drawer. Cloudy skies obscured the sun most of the next 3–4 days. For reasons he was unable to explain subsequently, he nevertheless removed the plates from the drawer and developed them on Sunday, 1 March 1896. He expected to find only faint images (*1,3,5,11*), but, to his astonishment, the images were as intense as those obtained previously when phosphorescence had been induced in the crystals by sunlight (*10,11*). Thus, irrelevant circumstances led *by chance* to his decisive observation (*4*).

Becquerel's "prepared mind" realized at once Nature was trying to tell him something, and there occurred a conscious recognition that the crystals were emitting penetrating radiations spontaneously, even in darkness. This *cognizance* of a new and unexpected atomic property of matter (*2*) constituted the discovery of radioactivity!

He published six papers concerned with his discovery in 1896 and two more early the next year (*6,7*). But, by the autumn of 1897, his attention turned to experiments with other physical phenomena, and it did not return to radioactivity until 1899. In that year he received some radium from Marie

* This thick paper envelope would have absorbed all α-particles. Almost all of the effects Becquerel observed photographically must have been due, then, to β-particles. The β-particles emitted with maximum energy of 3.3 MeV by 20-min Bismuth-214 are the most energetic of any of the radionuclides in the uranium series, and the maximum range of these would exceed 1.5 cm in water or other unit-density substances composed of low-Z atoms.

and Pierre Curie, with whom he had become acquainted.*

His discovery in 1899 that the radiations induced luminescence in diamond, zinc sulfide, and other substances (*3*) initiated a long chain of events leading to the evolution of our modern scintillation detectors. In this respect, then, his findings were the beginning of our present-day methods for "inside-out" in vivo studies of rates of accumulation, dynamic flow, and the distributions of γ-nuclides in our patients, as well as the use of the well-type scintillation detector for assays in vitro.

Becquerel's studies in 1900 led him to discover that the magnetically and electrostatically deviable β-particles of radium are negatively charged electrons emitted with high energies in a continuous distribution of velocities (*3,5*). In 1901 he showed that the spontaneous emission of radiations by uranium was not diminished at the temperature of liquid air (*5*). That year also he found that the radioactivity of uranium easily could be removed by dissolving barium chloride in a solution of uranium chloride and then adding sulfuric acid (*3*). The radioactivity (Crookes' Uranium X) seemed to become entrained with the barium sulfate precipitate. Eighteen months later, the lost radioactivity of the uranium grew in again spontaneously, whereas the precipitate lost its activity (*3,5*).

Becquerel also became interested inadvertently, by chance again, and in an intimately personal manner, in the physiologic effects of penetrating radiation from radium. While traveling to London to speak to The Royal Society (*10*), he carried a tube containing a large amount of radium in a pocket for about 6 hours. A publication with Pierre Curie in 1901 (*13*) disclosed his observation that, on arriving home in Paris, a reddened spot had appeared on his skin beneath the location of the vest pocket within which he had carried the tube 10 days previously. The glass tube was about 10–15 mm long and its diameter about 3 mm. The tube was wrapped in paper before putting it in a small cardboard box. The spot on the skin initially had the oblong shape of the tube. It became more deeply colored with time and took the shape of an oval about 6 cm long and 4 cm wide. Three weeks after the exposure through the glass tube, and through the box and his clothing, the skin became necrotic and the most affected part suppurated and sloughed. The ulcer was treated with applications of medicated dressings for a month. After the dead tissue was eliminated, the ulcer closed 7 weeks following the radiation exposure, but a scar remained (*2,13*). These unpleasant findings so upset Becquerel that he chided the Curies for their discovery of a substance which emitted radiation having such injurious effects (*10*). However, in his Nobel Lecture in 1903 (*2*) he stated that these effects were being explored for the treatment of cancer.

The new source of energy involved in spontaneous radioactivity persistently puzzled Becquerel. By the time of his presentation to The Royal Institution in London on 7 March 1902 (*10*), he attributed this energy to an association "with the disintegration of [radio]active matter." He thus anticipated, in part, the seminal hypothesis formulated that spring by Rutherford and Soddy of spontaneous atomic transmutation as the basic explanation of radioactivity (*5,7*).

Becquerel's discovery of radioactivity derived directly from his recognition that the autoradiographs, which he saw 80 years ago, were manifestations of a *new* phenomenon. This simple uncomplicated method of detection has been developed in the intervening decades until it provides the ultimate in radiographic resolution at the microscopic level. And this furnishes opportunities to do the elegant "atomic" histochemistry which now powerfully extends our methods to derive new knowledge at the cellular and even at the subcellular levels of tissue organization.

That Becquerel had immediate access to a particularly rapid means of publication, within 10 days after his discovery (*5,6*), may have been crucial to his becoming recognized as the discoverer of radioactivity. For Professor Sylvanus Thompson, a physicist experimenting in London, had found essentially simultaneously that uranium nitrate, when exposed to sunlight while resting on a shielded photographic plate, gave rise to radiations that blackened the developed emulsion beneath the location of the uranium salt. This so surprised him that he described the effect in a letter to the President of The Royal Society. The latter suggested in his reply on 29 February 1896 that Thompson publish his finding without delay. However, in a subsequent letter less than a week later, he informed Thompson that he was already too late, because Becquerel had just beaten him into print. Thus, Becquerel's opportunity to *publish with great speed* decisively established his priority. "Thompson's interest in the phenomenon cooled with the Frenchman's publication" (*6*).

It is interesting to speculate on what influences the conveniences stemming from the propinquity of Becquerel's home and his laboratory may have had on his making his discovery on a Sunday, as well as

* The discoveries of Polonium and Radium by the Curies in 1898 had stemmed from their having read Becquerel's publications (*7*).

upon his assiduous pursuit of his studies of it during the next dozen weeks. Becquerel's home was the professor's apartment* in The Museum of Natural History (*5,12*). Incidentally, he had been born there (*5*) on 15 December 1852 (*2,9*), during the time his father had held the same professorship.

Becquerel, like his father and grandfather before him, was elected to membership in The French Academy of Sciences. Many honors poured in upon him, naturally, including Fellowship in The Royal Society and membership in The Academy of Sciences in Washington. In 1903, he was awarded half of The Nobel Prize for Physics "in recognition of the extraordinary services he has rendered by his discovery of spontaneous radioactivity" (*2*). The other half was awarded to Pierre and Marie Curie "in recognition of the extraordinary services they have rendered by their joint researches on the radiation phenomena discovered by Professor Henri Becquerel" (*2*).

ACKNOWLEDGMENTS

Liliane and Robert Cohen, in Paris, gave invaluable assistance and obtained the photograph of Becquerel, made about the time of his discovery, from the Bibliothèque Nationale. Folke Knutsson, in Uppsala, kindly supplied the autoradiographic image and the picture Becquerel furnished of himself to The Nobel Foundation in 1903. Atis Freimanis translated the paper by Becquerel and Pierre Curie (*13*).

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3. GLASSTONE S: *Sourcebook on Atomic Energy*, 3rd ed. Princeton, N.J., Van Nostrand, 1967 [As the name implies, this is an encyclopedic collection of accounts of the circumstances and relationships of the basic discoveries related to atomic energy and of their consequences. It includes many good pictures of some of the discoverers]

4. KNUTSSON F: Becquerel and the discovery of radioactivity: *Acta Radiol [Diagn] (Stockh)* 16: 113–116, 1975 [A fine photograph of Becquerel is included, as well as the first autoradiographic image, observed by him in making his discovery on 1 March 1896]

5. ROMER A, ed: *The Discovery of Radioactivity and Transmutation.* New York, Dover, 1964 [Includes translations of five of Becquerel's publications, e.g., Refs. *1* and *8*]

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9. Antoine Henri Becquerel. In *World Who's Who in Science; A Biographical Dictionary of Notable Scientists from Antiquity to the Present.* Chicago, Marquis, 1968, p 141 [This is a splendid source of brief biographic sketches of most of the scientists involved in the origins of nuclear medicine]

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* Similarly, Röntgen's home was nearby, on the second floor, atop The Institute of Physics at The University of Würzburg. He had merely to descend to his laboratory on the floor below conveniently to indulge himself in experimentation at odd times with his electrically excited evacuated Hittorf–Crookes discharge tubes. These endeavors, born of an enthusiastic curiosity, resulted in the serendipitous discovery in Röntgen's "prepared mind" during the night of 8 November 1895 of "the rays named after him."

These monumental discoveries, made less than 4 months apart, were pivotal to the many fundamental discoveries in the physical and chemical sciences stemming from them and from which the basic aspects of nuclear medicine have emerged. Perhaps accelerations in the rates of scientific creativity might become facilitated were intuitive and dedicated experimentalists, endowed with scholarly imaginations and having well-trained minds, again to be furnished with homes in proximity to their laboratories.

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