2.2.2 Radioactivity - Historical Background

> *To the chemists of the 19th century the atom and the element represented each in its sphere the uttermost limit of chemical subdivision or disintegration, and at the same time the point beyond which it was impossible for experimental investigation to proceed. If it were queried what there was beyond, nothing but more or less vague and fruitless speculations were forthcoming. This line of demarcation, for so long regarded as insurmountable, has now been swept away, at all events in principle. Nowadays the inner structure of atoms and the laws regulating that structure belong to the problems that can be made the subject of discussion in a thoroughly practical and at the same time fully scientific manner, thanks to the exactness of the measurements which have been taken. The results already arrived at are not only of the utmost importance in themselves, but derive perhaps a still greater significance from the numerous possibilities, wholly unsuspected ten or twelve years ago, which have been thrown open for the continuance of the work of investigation in this department of science.* — Presentation of 1908 Nobel Prize in Chemistry to Ernest Rutherford

On March 1, 1896, Antoine Henri Becquerel discovered radioactivity.[40] He was looking for X-rays (recently discovered in November 1895 by Roentgen) from phosphorescent materials, mainly uranium. His motivation was to look for X-rays (recently discovered by Roentgen) from phosphorescent materials, and he was familiar with the phosphorescent properties of uranic salts,[41] which, of course, contain uranium. He wrapped a photographic plate (a piece of glass covered with a photographic emulsion) in black paper, and placed on the paper a piece of a phosphorescent substance. He exposed the combination to the sun for several hours, in the expectation that the sunlight would cause the uranium to phosphoresce, and that phosphorescent light from the uranium would penetrate the black paper and leave an image on the emulsion. It worked as expected, but then there came a week of cloudy weather and the sun did not shine. Becquerel put his plates and uranium in the cupboard for a week (without being exposed to sunlight), and for some reason he decided to develop those plates, even though he expected nothing. However, his intuition was correct, and he discovered that the plates showed an image, just as if it had been in the sun!

This was the first step in the discovery and understanding of radioactivity, and the "rays" that must have been emanating from the uranium were called "Becquerel rays."

One week later, on March 9, Becquerel discovered that the rays could discharge an electroscope, which meant that the rays were charged. At that time there were two types of rays known, cathode rays (which had been shown by Thomson to be electrons) and light rays (which had been shown by Maxwell and Hertz to be electromagnetic waves). Of course, the "X-rays" of Roentgen would turn out to be high-frequency electromagnetic waves, and the Becquerel rays were nothing but electrons, but that was not clear for quite a while. In fact, the uranium sample emitted both electrons and α-particles, but the α particles were

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[40] This history is taken primarily from *Inward Bound* by Abraham Pais.

[41] Specifically, Becquerel used uranyl disulfate, $K_2UO_2(SO_4)_2 2H_2O$.

easily stopped by the paper and so did not contribute to the darkening of the emulsion.[42]

The second step in the understanding of radioactivity came in 1898 when Marie and Pierre Curie found that the element thorium ($Z = 90$) was also radioactive. In addition, they discovered two new elements due to their radioactivity, polonium and radium. These latter two they found by chemically isolating them from their sample of pitchblende. Pitchblende is a black mineral, mainly UO_2, but it also has some impurities, and these are what the Curies found. The Curies won the 1903 Nobel Prize in Physics, jointly with Becquerel, for their investigation into radioactivity. Marie won the 1911 Nobel Prize in Chemistry for the discovery of polonium and radium. At this point, even though radioactivity was not at all understood yet, two questions had been become common: Where did the energy associated with the activity come from? and: Were *all* elements radioactive (but perhaps with very long lifetimes)?

The third event in our story ocurred in 1899 when Rutherford deduced that there were two different types of Becquerel rays: α rays and β rays. They were distinguished by their ability to penetrate matter: α rays were easily absorbed in a few centimeters of air (Becquerel's black paper absorbed them); β rays were more penetrating—it took several cm of air before they were absorbed. Later it was determined that α rays were actually the nuclei of ^{4}He, and β rays were electrons. In 1900, Paul Villard in Paris observed a third type of ray emitted by radium that was even more penetrating than β rays (but it was not charged), and he called them γ rays. These, of course, were photons, but that was not determined until 1914.

The final piece of the puzzle, the fourth step, was put in place in 1902 when Rutherford and Frederick Soddy [Nobel Prize, Physics, 1921] developed their "transformation theory." This theory was an explanation of what was occurring during radioactive decay: In modern terminology, a "parent" nucleus was transformed into a "daughter" nucleus when an α or β ray was emitted. Soddy had originally suggested the term "transmutation theory," but Rutherford objected, believing that people would think they were proposing medieval alchemy. In fact, though, that was exactly what they were doing: radioactivity was changing one element into another. Another part of the transformation theory was the observation that the process of transformation decayed exponentially with time. They discovered this while investigating a gas called "thorium emanation," which we now know was an isotope of radon, ^{220}Rn. Most of the daughter elements were solids at room temperature, so that they remained locked in the original rock. Radon, however, is a gas, and so when it is created as a part of a series of radioactive decays it can be easily isolated. Rutherford and Soddy found that no matter when they started observing, the activity of ^{220}Rn was reduced by half in one minute, and this allowed them to describe radioactivity mathematically as an exponential decay.

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[42] Becquerel's family was quite prodigious. Along with his grandfather, Antoine César, his father, Alexandre Edmond, and his son, Jean, the four of them continuously held the chair of physics at the Museum of Natural History in Paris from 1838-1948, a span of 110 years! The four of them studied many aspects of physics, including thermoelectric phenomena, luminescence, infrared spectroscopy, magnetic polarization by crystals, and magneto-optics. In fact, after his discoveries, Antoine Henri said, "These discoveries are only the lineal descendants of those of my father and grandfather on phosphorescence, and without them my own discoveries would have been impossible."