No. 1 in a series of essays on radioactivity produced by the Royal Society of Chemistry Radiochemistry Group

Discovery of Radioactivity

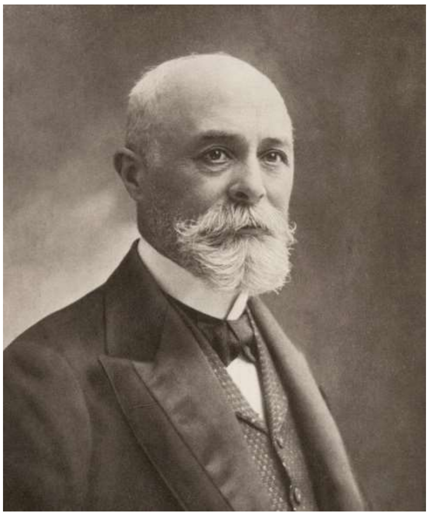
Earth has been radioactive since it was formed 4500 million years ago. In fact the age of the earth can be calculated from a detailed examination of its radioactivity, particularly the decay of uranium to lead.

Henri Becquerel

Following the discovery of x-rays in 1895 by Roentgen, Becquerel had the idea, mistakenly, that minerals that were made phosphorescent by visible light might emit x-rays. (X-rays are still used to look for faults such as breaks and cavities. X-rays, not absorbed by materials or the specimen, can be detected).

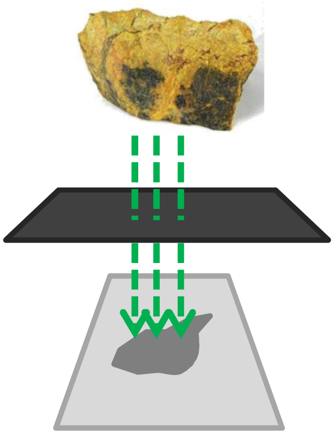
Phosphorescence is the ability of a crystal to absorb light and re-emit light sometime after the exciting light has been removed. The Romans used phosphorescent minerals in the centre of some of their roads to illuminate them at night. The minerals were stimulated by daylight then continued to re-emit light during the hours of darkness.

Being a mineralogist, Becquerel had a large collection of minerals, many of which exhibited phosphorescence. He experimented by wrapping a photographic plate in black paper (to protect it from direct light), placing a phosphorescent uranium mineral on it and exposing it to bright sunlight. When developed the plate bore a clear image of the uranium mineral. Initially, he thought of this as confirmation of his theory.



Henri Becquerel

In February 1896 there was very little sunlight in Paris for several days (a common occurrence at that time of year). Due to this, Becquerel stopped his experiments and placed the uranium mineral and wrapped photographic plate together in a drawer. After several days he developed the plate expecting a weak image from the small amount of phosphorescence that had not yet decayed away.



Radioactivity blackens a photographic plate even through a layer of black paper

Imagine his surprise to find the image as intense as in the original sunlight experiment. He now drew the correct conclusion that this had nothing to do with light but the exposure came from uranium itself, even in the dark. Radioactivity had been discovered. He went on to show that uranium minerals were the only phosphorescent minerals to show this effect.

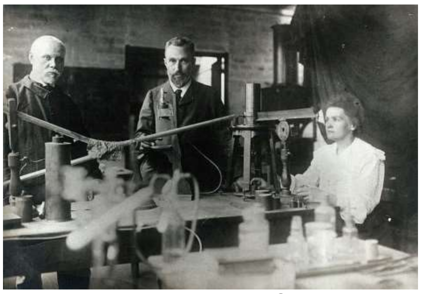
The Curies

Becquerel recommended that Marie Sklodowska Curie choose the subject for her doctoral thesis. During her studies she quickly established that the amount of radioactivity emitted by a uranium mineral was proportional to the amount of uranium present in it. Further, that thorium minerals behaved similarly.

Marie Curie

When she examined the uranium ore pitchblende she found that it was four times more radioactive than would be expected from the uranium mineral alone. The only conclusion was that it contained another unknown radioactive component.

With the help of her husband Pierre they boiled 40 pound (20 kilo) batches of pitchblende in cast iron cauldrons and in 1898 they succeeded in isolating two new radioactive elements.



Pierre (centre) and Marie Curie in their laboratory

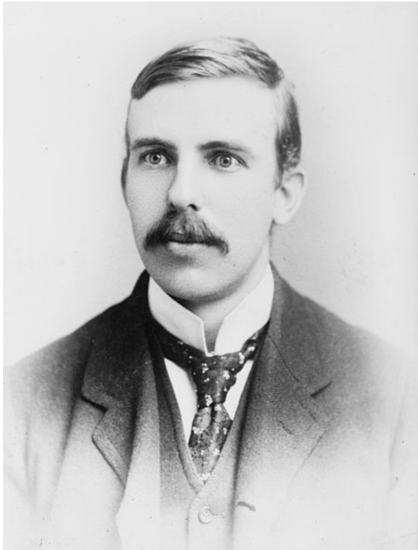
The first they called Polonium (Po), after Marie's native Poland, and the second Radium (Ra) after the Latin for ray (radius). Po was 60 and Ra 400 times more radioactive than uranium (U). This was the invention of radiochemistry. In radiochemistry the destination of any species in physical or chemical separations is monitored by their radioactivity. Becquerel and the Curies received the 1903 Nobel Prize in Physics for these experiments.

During this period the Curies did ALL the heavy work themselves on Pierre's income of £20/month. For this he gave 120 lectures/year. By 1902 they had produced about 0.1 g of pure radium chloride (about 70mCi, millicuries, or 2600 MBq, megabecquerel).

Rutherford, Royds and Soddy

Ionization is the removal of one, or more, electrons from an atom, leaving it positively charged. In studying the ionization of gases by uranium, Rutherford identified two types of radiation: α-radiation, which produces by far the greater part of the ionization which can be stopped by a single sheet of paper, and β-radiation, which produces much less ionization and is capable of penetrating several millimetres of Al, Cu, etc. In 1900 γ-radiation was discovered by Villard who found that radium emitted an extremely penetrating radiation capable of detection through 20 cm of iron or several cm of lead. γ-radiation is electromagnetic radiation like light but the photons have much shorter wavelengths and higher energies.

Many experiments were carried out with α radiation, usually from radium prepared by the Curies, which suggested that the α particle was a helium nucleus. The definitive identity of α-radiation was established by Rutherford and Royds in an elegant experiment. They put a large quantity of purified radon gas into a thin-walled glass tube.

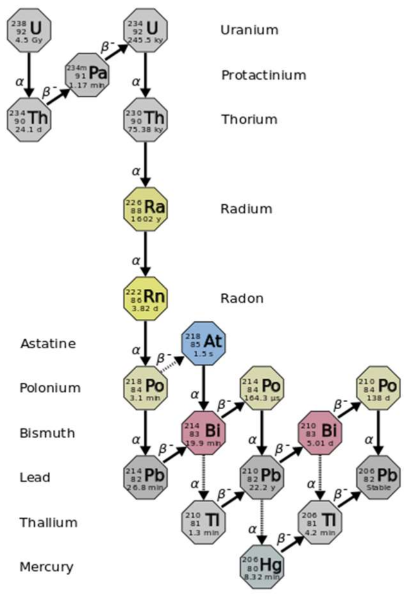


Ernest Rutherford

The wall of this tube was thin enough to allow most of the α-particles through. An evacuated collecting tube surrounded the thin-walled tube containing the radon. After two days the yellow helium line was clearly visible in the outer tube by visible light spectroscopy. Then after six days the whole spectrum of helium was visible. The α-particles thus gave rise to the observed helium. Identifying helium in this way is like recognizing a neon light from its colour.

Uranium Decay Series

When uranium (U) and thorium (Th) emitted α and β particles they decayed to different elements, this sequence was called the natural decay series. By studying the chemistry of the natural decay series of uranium and thorium 30 new radioactive elements were discovered, of which radium (Ra) was the best known. This was a truly international effort but much of the work was done by Rutherford and Soddy who both obtained Nobel prizes in Chemistry for this work.



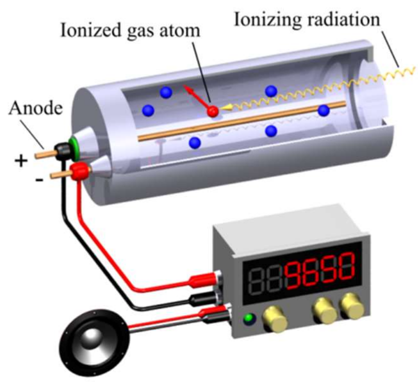
Unravelling this chemistry was like the work of a detective. Many of these "elements" could not be separated by chemical methods, e.g. radiothorium (^{228}Th) from Th, Ra from mesothorium and radium D (Ra-D = ^{210}Pb) from the final product in the decay chain, lead (Pb). Soddy concluded that such inseparable "elements" must occupy the same position in the Periodic Table and called them "isotopes". Isotopes are atoms of the same nuclear charge but different mass; that is they contain the same number of protons (and are the same element) but different numbers of neutrons.

This concept was confirmed in the early 1920s when Aston, using the first mass spectrometer (a device which uses a magnetic field to separate atoms of different mass), was able to separate and measure isotopes for a number of elements, in particular ^{20}Ne and ^{22}Ne. The placement of these new elements in the periodic table in 1913 was remarkably accurate when compared to the present allocation.

From the detailed chemistry of the decay series Soddy and others noticed that the chemistry of the daughter element was connected with the emitted radiation. It eventually dawned on the researchers that α particle emission moved the element two places down the Periodic Table, whereas β emission moved it one place up the Table.

Measurement

Having identified the various types of radioactivity it was necessary to be able to measure them.
Rutherford's doctoral thesis at the Cavendish Laboratory under J.J. Thompson in 1895 involved studying the effects of x-rays on gases. This later extended to radioactivity.



Geiger-Muller Tube for the detection of ionizing radiation

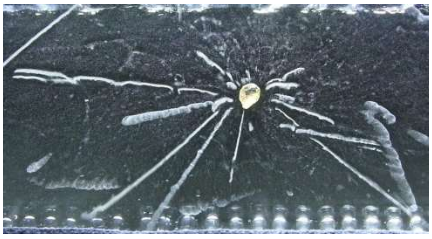
The work led to a clear understanding of the transport of electricity through gases by charged ions being attracted to the electrodes. This production of charged ions was used for detecting radioactivity in ionization chambers and Geiger-Muller counters.

Sir William Crookes (1832-1919) had a private laboratory at his home in Kensington Gardens, London. Here he discovered thallium (Tl), and the spintharoscope. The latter being a method of quantitatively detecting radioactivity by eye from the light flashes produced on a zinc sulphide (ZnS) screen. The light flash caused by radioactivity is a scintillation event. The spintharoscope was widely used by the early researchers, including Rutherford's group when discovering the nucleus. In addition, spintharoscopes were sold to the general public for "after dinner" entertainment in well-to-do houses.

Curie Electrometer

The electrometer was used to determine the presence of radioactivity in solutions and precipitates. It was used by the Curies to measure the radioactivity of their solutions when separating polonium and radium from pitchblende. Electroscopes consist of two light gold leaves that, when charged, spring apart. When radioactivity passes through the electroscope the ionization causes the charge to leak away and the gold leaves to collapse together at a rate proportional to the number of ions produced, which, in turn, can be related to the amount of radioactivity. Modern versions are often a metal-coated quartz fibre and are used as personal dosimeters.

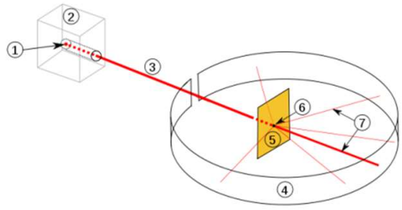
The cloud chamber was invented by C.T.R. Wilson after studying the meteorological effects of clouds on Ben Nevis. In effect, the track of an ionizing particle could be photographed as it made a "cloud trail" in a chamber saturated with vapour. The principle is similar to the formation of aircraft trails at high altitudes. Cloud chambers are still used today in many particle physics experiments.



Thorite mineral in a cloud chamber. The streaks are 'clouds', nucleated by radiation

Discovery of the Nucleus

In 1911 Geiger and Marsden investigated the outcome of bombarding various materials with α particles. While bombarding gold foils, it was noticed that very occasionally one particle would be scattered through an angle greater than 90°. Rutherford was heard to say; "It was quite the most incredible event that has ever happened to me in my life. It was almost as incredible as if you had fired a 15-inch shell at a piece of tissue paper and it came back and hit you." Rutherford drew the conclusion that the major part of the mass of an atom must be concentrated in a central nucleus and from the scattering experiments he calculated that the size of the nucleus must be about 10^{-14}m, very small when compared to the approximate size of an atom (10^{-10}m).



Rutherford's scattering experiment. 1) alpha source, 2) lead shielding, 3) alpha beam, 4) fluorescent detector, 5) gold foil, 6) target, 7) some particles pass through, some are deflected

Assuming a nucleus to be the size of a balloon, then the outside of the atom would be about 3km away. A drop of water made of atoms that size would occupy the whole galaxy. Further α bombardment experiments were carried out by Rutherford's team, firstly on hydrogen and then later on nitrogen gas. In both cases, protons were produced with very high energies. The conclusion was that a very few nitrogen atoms had been transformed into oxygen atoms. This was confirmed by photographing tracks in a Wilson cloud chamber. Thus nitrogen can be transmuted (transformed from one element to another), on a minute scale, to a stable isotope of oxygen. The alchemists dream of transmutation had come true. This gave rise to intense interest in experiments in which one nucleus was bombarded with another.

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

Transmutation of ^{14}N by an α particle to form a proton and ^{17}O

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