The Discovery of Artificial Radioactivity

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Frédéric Joliot was born in Paris on 19th March 1900. His older brother was killed in the first few days of the 1914-1918 war. Frédéric joined the *École de physique et chimie industrielles* in Paris, where he studied physics at the top of his class. In December 1924, director of studies, Paul Langevin, presented him to Marie Curie; he became her personal assistant at the Radium Institute, which was created for her in 1909 by the University of Paris and the Pasteur Institute. Marie Curie assigned her daughter, Irène, then her assistant, to guide the young man.

Frédéric and Irène married in 1926. Shortly after, they decided to carry out experiments together; Irène would continue to sign their publications under her maiden name. They prepared very intense sources of polonium on very small surfaces, thanks to the Radium D[1] stocks accumulated by Marie Curie: the alpha particles[2] emitted by radioactive polonium served as projectiles to explore nuclei.

The Legacy of Marie and Pierre Curie

Of course, Irène and Frédéric, in their respective Nobel speeches, paid homage to Pierre and Marie Curie, Irène's parents. It is particularly thanks to Marie Curie that they had significant sources of radioactive bodies such as polonium, produced from Radium D, coming from light bulbs which contained radon for medical usage, which Marie Curie had carefully stored at the Radium Institute.
Frédéric Joliot in particular made a very moving tribute to Marie Curie (though she had died in July 1934 she still had had the time to witness, in the January of that year, the discovery of artificial radioactivity carried out by her daughter and her son-in-law):

It was certainly a great satisfaction for our late teacher Marie Curie to have seen extend this list of radioelements which she had had, together with Pierre Curie, the glory to introduce.

1. The elements of the radioactive family of radium were still not all identified, also bearing this type of name (Radium D is an isotope of lead).

2. The alpha particles $^{4}_{2}He$ are helium nuclei (without electrons), consisting of two protons and two neutrons. Frédéric Joliot called them "*helions*" in his text.

In 1930, very penetrating radiation had been observed in Berlin during the bombardment of light nuclei with alpha particles[3]. In January 1932, Frédéric and Irène highlighted the fact that this same radiation – which they thought was comprised of gamma rays – has the ability to eject protons of hydrogenated substances (such as wax or cellophane). Based on their results, a month later, British man James Chadwick showed that it is actually "neutrons". In 1934, the young couple discovered artificial radioactivity - mentioned a little later - which won them the Nobel Prize in Chemistry.

Figure 1: ***Frédéric and Irène Joliot-Curie in their laboratory in the Radium Institute in 1934*** *(Photograph: Association Curie-Joliot Curie ACJC)*

Frédéric was appointed in 1937 professor at the *College de France* and became director of the establishment's Nuclear Chemistry Laboratory, as well as the Atomic Synthesis Laboratory in Ivry of the National Fund for Scientific Research, the precursor of the CNRS. Irène, appointed in 1936 professor at the Faculty of Science at the University of Paris, became for a few months the first Under-Secretary of State for scientific research in Léon Blum's government.

In December 1938, Otto Hahn and Fritz Strassmann discovered the fission of uranium under the action of neutrons, the physical explanation of which would be given by Lise Meitner and Otto Frisch. Frédéric Joliot immediately launches

3. This experiment was done in Berlin-Charlottenburg by Walter Bothe and Herbert Beker, by the bombardment of beryllium or boron by the α particles from a source of polonium. At the time (1920 to 1932, before the discovery of the neutron), it was thought that the nuclei were made of protons, α particles and electrons. Heisenberg reported to the 1933 Solvay Conference that "the discovery of the neutron by Curie and Joliot and by Chadwick (...) shows beyond doubt that these neutrons may appear as independent components of the nucleus, beside protons and α particles".

into the study of this new phenomenon: he confirms its reality thanks to the enormous energy released; then with his co-workers, Hans Halban and Lew Kowarski, he studies the possibility of a chain reaction. A remarkable series of experiments were carried out and calculations of an "energy producing device" were made with the help of Francis Perrin. Patent applications were filed in early May 1939; Frédéric contacted the Mining Union of Haut-Katanga – which put 8 tons of uranium oxide at its disposal - in order to achieve a first atomic pile [nuclear reactor] and to obtain energy for industrial use. In September 1939, the war disrupted this programme, but the work continues. The team showed that the device must be heterogeneous, the uranium immersed in a moderator that should be heavy water, and that an enrichment of uranium 235 would be favourable; the Danish physicist Niels Bohr had shown early in 1939 that it is the rare isotope of uranium, uranium 235, which undergoes the fission by slow neutrons. A French special commando managed to bring heavy water from Norway which was occupied by the Germans.

However, in May-June 1940, the German invasion of France required retreat into Clermont-Ferrand, then Bordeaux: Halban and Kowarski were sent with heavy water to England, the uranium hidden in Morocco. Joliot decided to stay in France. He found his laboratory occupied by the Germans, but placed under the authority of a physicist friend, Wolfgang Gentner, who had worked for two years at the Radium Institute of the still alive Marie Curie, and who directly witnessed the discovery of artificial radioactivity; as an anti-Nazi, Gentner effectively protected Frédéric. He was actively involved in the Resistance and adhered to the Communist Party.

At the Liberation, Joliot was appointed general director of the CNRS. General de Gaulle created the Commission for Atomic Energy, of which Frédéric Joliot became the first High Commissioner in 1945. The first French atomic pile (nuclear reactor), Zoé, came into operation in December 1948. However, the scholar actively campaigned in the international peace movement and he launched in March 1950 the famous "Stockholm Appeal" to ban nuclear weapons. A month later, during the "Cold War", his political position earned him dismissal from his post as High Commissioner. In 1956, Irène Joliot-Curie died suddenly, after having established plans for a new laboratory in Orsay. Frédéric followed suit; he organised the new institute, equipped with a synchrocyclotron. But he in turn died prematurely in August 1958. General de Gaulle decreed a state funeral.

Figure 2: ***Frédéric Joliot in 1947 dans in his CEA office*** *(photograph: Association Curie-Joliot Curie ACJC).*

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Let's go back to 1934. In January of that year, Frédéric and Irène Joliot-Curie discovered that positron emission (thus calling the electrons positive, by analogy with "electrons") which they observed in the bombing of a sheet of aluminium by the alpha particles of polonium is not instantaneous, but decreases exponentially in minutes. They had just discovered "artificial radioactivity": they formed in the aluminium a radioactive phosphorus, previously unknown, a stable isotope phosphorus.[4] Then they highlighted two other artificial radioelements. A fortnight later, chemical separations, conducted in a very short time, confirmed their interpretation. This was a remarkable generalisation of the phenomenon of radioactivity that would rapidly have significant consequences. The discovery would also merit the young scholars to win the Nobel Prize in Chemistry 1935.

The Positive Electron or "Positron"

The positive electron, also known as positron, was discovered in 1932 in cosmic rays by American physicist Carl D. Anderson (1905-1991, Nobel Prize in Physics 1936). It was immediately considered as

4. The reaction is given by F. Joliot in his text $^{27}_{13}Al + ^{4}_{2}He \rightarrow ^{30}_{15}P + ^{1}_{0}n$; from the aluminium, an unknown isotope of phosphorus is obtained $^{31}_{15}P$ (the isotope has 15 neutrons and not 16). This isotope is radioactive and does not exist in nature, having been disappeared for a long time.

"the antiparticle" of the electron, predicted by the English theoretical physicist P. A. M. Dirac. The absorption in the field of gamma radiation over 1.02 MeV can give birth, by "materialisation", to an electron-positron pair. Conversely a positron, at the end of the process, and an electron, when they meet, "annihilate", the energy corresponding to their masses appearing as two photons, of 0.51 MeV each, issued back-to-back, which was verified experimentally by F. Joliot in 1933. Two of the new radioelements discovered by F. and I. Joliot-Curie in 1934 decreased by the positron emission (the first observation of radioactivity β^+)[5].

Confirmations came quickly from foreign laboratories which employed deuterons or protons as projectiles: from Berkeley, in California, where Ernest Lawrence and his co-workers realised that several pieces of their cyclotron had become radioactive without their knowing, from Cambridge, in England, where John Cockcroft and his co-workers produced radioactive nitrogen from carbon, using their own accelerator.

The production of new radioelements by charged particles was at the time limited to light elements, such as boron or aluminium. In Rome, Enrico Fermi had the idea of using neutrons which, not being charged, could penetrate the heaviest nuclei. With his team, he thus irradiated all the elements that he could get a hold of. He immediately had positive results. From June 1934, Fermi was able to declare his discovery of new radioactivity in 47 elements out of the 68 elements studied so far. The team in Rome observed several new radioactivities by bombarding the heaviest element, uranium, with neutrons; they thought that they had formed "transuranium elements" which do not exist on Earth. Are there any elements beyond uranium? These studies would be taken up in Berlin by Lise Meitner[6] and Otto Hahn, then by Irène Joliot-Curie and Paul Savitch in Paris.

George de Hevesy (1885-1966, Nobel Prize in Chemistry 1943), Swedish chemist of Hungarian origin, had shown in 1913 that radium D was a radioactive isotope of lead (lead 210, beta emitter, with a period of 22 years, located in the radioactive family of radium). He imagined then that we could trace the lead

5. To return to the reaction of F. Joliot in note 4 above, the radioactive isotope of phosphorus will be transmuted by a reaction of radioactivity β^+ (positron emission), that the Joliot pair mention in their note from January 1935 to the *Académie des sciences*: *"The isotope* $^{30}_{15}P$ *of phosphorus would be radioactive with a period of 3mn15s and would emit positive electrons following the reaction* $^{30}_{15}P \rightarrow {}^{30}_{14}Si + e^+$ *"*. Thus two new phenomena that the Joliots discovered simultaneously: radioactivity caused artificially, that of the isotope of phosphorus, and the reaction of radioactivity by positron emission (which will be called radioactivity β^+).
6. In 1938, the "Anschluss" of Austria compelled Lise Meitner, of Jewish origin, to emigration.

thanks to its radioactive isotope, which was called a tracer or indicator. Lead, however, is not a constituent element of living organisms, having no biological applications. But, in September 1935, in Copenhagen, Hevesy and his colleague Otto Chiewitz used, for the first time, an artificial radioelement, phosphorus 32, as a radioactive tracer for the study, in rats, of phosphorus metabolism.

Radioactive Tracers in Biology and Medicine

Some radioactive isotopes (known as tracers or indicators) are added to the stable isotopes of an element. The radioactive isotope will behave, chemically and biologically, like stable isotopes. It may be included in an organic molecule (labelled molecule). It is then possible, thanks to its radiation, to study in a living body the metabolism (fixation, elimination...) of the element considered. In medicine, tracers are used for diagnosis. Like, for example, iodine-123 (period 13h) and particularly iodine-131 (period 8j) in the thyroid, the technetium-99m in the lung. Also invented is "positron emission tomography" (PET) which makes great service. Thus introduced into the organism is water labelled at oxygen 15 (period of 2 minutes only); oxygen 15 is a positron emitter; these will, as has already been said, be destroyed at the end of the process with electrons and each time will give two gamma rays emitted back-to-back, which will be detected coincidentally by the detectors of a "positron camera".

It is necessary to distinguish the physical periods (such as those indicated above) from the biological periods (taking into account the natural elimination of substances).

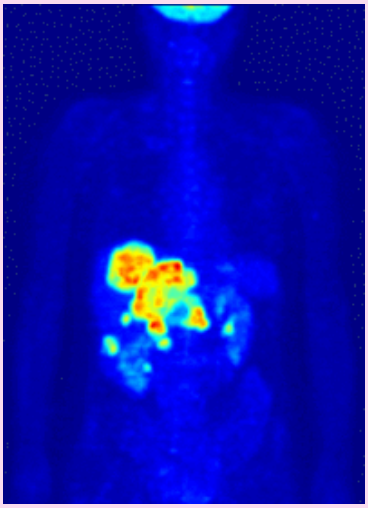

***Figure 3:* Positron emission tomography.** *An artificial radioactive element, Fluorine 18 (one neutron less than the classic Fluorine 19) is incorporated into a glucose solution. Cancerous cells, with high activity, need energy in the form of glucose and stabilise the solution. The positrons*

emitted by radioactivity β^+ by fluorine will be destroyed with electrons and will emit photons γ, which are visible on the photograph, thus making it possible to locate the cancerous cells.

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The Nobel 1935 Prize award ceremony took place in Stockholm on 12th December. Frédéric and Irène recognised some works carried out abroad following their discovery. They decided to hold two lectures: Irène, often considered as the chemist of the team, hosted physics, while Frédéric, considered the physicist, hosted chemistry!

***Figure 4:* Frédéric and Irène Joliot-Curie during the ceremony of the Nobel Prize award ceremony, in Stockholm, 12th December 1935** *(photograph: Curie-Joliot Curie ACJC).*

Frédéric Joliot actually divided his presentation into two parts. The first was devoted to the description of chemical experiments that had allowed the researcher couple to separate and identify the new radioactive elements. Joliot emphasised the existing difficulty in characterising these elements, given the small masses at play:

> *If we can write with certainty the nuclear reaction corresponding to the majority of spontaneous transmutations, it is not the same for induced transmutations. The productivity of these transmutations is very small and the element masses formed by using the most intense projectile sources that we can currently produce are less than 10-15g, represented by no more than a few million atoms.*

Among these new elements, appears the radioactive phosphorus formed in aluminium, for which Joliot gives the production reaction:

In the case, for example, where aluminium irradiated by alpha rays emits neutrons, the preceding rule makes it possible to write the next transmutation reaction:

$$^{27}_{13}Al + ^{4}_{2}He \rightarrow ^{30}_{15}P + ^{1}_{0}n$$

With the atom formed being radioactive, we can verify that it has the chemical properties of phosphorus.

The radioactivity of phosphorus 30 leads to the emission of positive electrons, readily detected through the thin wall of the glass tube, provided that it moves quickly. Here was also the first observation of radioactivity β^+. This radioactivity can be written as:

$$^{30}P \rightarrow {}^{30}Si + e^+ + \nu$$

where ^{30}Si is a stable nucleus and ν represents a neutrino.

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In the second part of his lecture, Frédéric explains how "these experiments were taken and developed in several countries [...] using projectiles protons and deuterons", provided by installations of very high voltages, as well as with neutrons. He adds:

Currently, we are able to synthesise [...] *more than fifty new radioelements, a number already exceeding that of natural radioelements that are found in the earth's crust.*

However, despite efforts already underway, new particle accelerators are absolutely necessary:

The diversity of the chemical natures, the diversity of average lives of these synthetic radioelements will make possible without a doubt new research in biology and in chemical physics. To carry out this work well, it will be necessary to have relatively significant amounts of these radioelements. It is by using artificially accelerated projectiles that this goal will be reached. [...] *Until then, only elements with a relatively short life, ranging from fractions of a second to a few months, have been obtained. In order to create a significant amount of an element*[7] *with a much longer life, you would need to have a source of supremely intense projectiles. Is there no hope in realising this new dream?*

To the wide range of particle accelerators – notably making it possible to produce radioactive bodies in significant amounts – would be added, some years

7. F. Joliot refers to the radioactive isotopes as *radioelements* and sometimes simply as *elements*.

later, nuclear reactors (atomic piles) which provide large amounts of neutrons, and which are also capable of producing in plenty certain radioelements.

The scholar then develops, in a flamboyant, daring and in part premonitory manner, his vision of the different possible applications of artificial radioactivity. The necessary amounts of radioelements depend on the applications considered. First and foremost, biology and medicine:

> *The method of radioactive indicators previously reserved for elements with raised atomic masses can be generalised to a very large number of elements distributed throughout the entire periodic table. In biology, for example, the indicator method, using synthetic radioelements, makes it possible to study more easily the problem of locating and eliminating various elements which are introduced into living organisms. In this case, radioactivity is only used to determine the presence of an element in some region or another of the organism. It is not useful in these studies to introduce significant amounts of the radioactive element.* […] *In the places, which we will learn to better understand, where the radioelements will be located, the radiation that they emit will produce its effect on neighbouring cells. For this second instruction for use, it will be necessary to use large amounts of radioelements. This will likely find a practical application in medicine.*

The first instruction for use indicated in this quotation corresponds to biological studies and diagnostic uses in medicine; the second instruction for use corresponds to medical treatment (Brachytherapy). The applications in biology and in medicine would indeed be numerous and lead to fundamental results.

Frédéric Joliot then turned to astrophysics. This subject also has a story:

> *Of all the facts considered, we understand that the few hundred atoms of different species that make up our planet should not be considered as having been created once for all and for ever. We observe them because they survived. Others less stable disappeared. These are probably some of the disappeared atoms that are regenerated in the laboratory.*

The scholar had suspected the possibility of chain reactions which could produce energy on a large scale. Thus the following sentence from his lecture is often cited:

> *If, turning to the past, we look back on the progress achieved by science at an ever-increasing pace, we have a right to believe that the researchers either building or breaking elements without limit will be able to carry out transmutations of an explosive character, real chemical chain reactions.*

As we indicated above, such a chain reaction would not actually be carried out or highlighted until 1939. F. Joliot concludes his presentation by returning to astrophysics:

> *Sometimes astronomers observe that a star of mediocre brightness suddenly increases in size, a star invisible to the naked eye can become very bright and visible without instruments, the appearance of a Nova. This sudden blaze from the star is perhaps caused by these transmutations of an explosive character,[8] a process that researchers will without doubt seek to achieve by hopefully taking the necessary precautions.*

Here the reader will see an evocation of thermonuclear fusion reactions, which we did begin to consider, but whose nature will not be explained or studied until a little later on. The implementation of some of these reactions is currently based on a major international project of power generation (Project ITER).

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(April 2009)

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8. Here Frédéric Joliot had, before even the consolidation of the hypothesis that stars are formed by nuclear reactions with a release of energy, a premonitory vision of the explosive birth of supernovae.

RÉPUBLIQUE FRANÇAISE

MINISTÈRE
DE L'INDUSTRIE ET DU COMMERCE

SERVICE
de la PROPRIÉTÉ INDUSTRIELLE

BREVET D'INVENTION

Gr. 5. — Cl. 2.

N° 976.541

Dispositif de production d'énergie.

CAISSE NATIONALE DE LA RECHERCHE SCIENTIFIQUE résidant en France (Seine).

Demandé le 1er mai 1939, à 16h 55m, à Paris.
Délivré le 1er novembre 1950. — Publié le 19 mars 1951.

(Brevet d'invention dont la délivrance a été ajournée en exécution de l'article 11, § 7, de la loi du 5 juillet 1844 modifiée par la loi du 7 avril 1902:)

On sait que l'absorption d'un neutron par un noyau d uranium peut provoquer la rupture de ce dernier avec degagement d énergie et émission de nouveaux neutrons en nombre en moyenne supérieur à l'unité. Parmi les neutrons ainsi émis, un certain nombre peuvent à leur tour provoquer — sur des noyaux d'uranium — de nouvelles ruptures, et les ruptures de noyaux d'uranium pourront ainsi aller en croissant suivant une progression géométrique, avec dégagement de quantités extrêmement considérables d énergie.

On s'est rendu compte, conformément à la présente invention, que si l'on parvenait à provoquer une telle réaction au sein d'une masse limitée d'uranium (ou de composé d'uranium ou d'un mélange contenant de l'uranium), on pourrait extraire de cette masse et utiliser à des fins industrielles l'énergie ainsi développée par les chaînes de ruptures successives.

Mais on se heurte immédiatement à une difficulté primordiale : ces chaînes pouvant se ramifier d'une manière illimitée, la réaction peut devenir explosive, ce qui restreindrait considérablement les possibilités d'utilisation de la masse d'uranium en question comme source maniable d'énergie industrielle.

On a donc cherché à maîtriser le dégagement d'énergie en l'empêchant de devenir explosif, et l'on a eu l'idée à cet effet :

1° Tout d'abord de réduire la vitesse de tout ou partie des neutrons libérés, de telle sorte qu'ils deviennent des neutrons lents, approximativement en équilibre thermique avec le milieu.

Cette réduction de vitesse donne déjà un moyen de stabilisation par le fait que la probabilité qu'a un neutron de quitter le dispositif avant d'être absorbé augmente avec la température. Elle permet d'autre part de réaliser des conditions dans lesquelles l'élévation de température amène rapidement un changement des concentrations ou des répartitions des constituants du dispositif, arrêtant ainsi le développement des chaînes;

2° Ensuite, d'introduire dans le dispositif un ou plusieurs éléments — à l'état de corps simples ou de combinaisons chimiques — susceptibles d'absorber les neutrons ralentis en proportion d'autant plus forte par rapport à l'absorption par l'uranium que la température est plus élevée.

Par le premier de ces moyens ou l'emploi conjugué de ces deux moyens, les chaînes peuvent se développer jusqu'à ce qu'une énergie suffisamment importante soit libérée, et être alors automatiquement interrompues ou limitées, évitant ainsi le développement explosif de la réaction.

On arrive ainsi à libérer de la masse d'uranium considérée, en vue de son utilisation industrielle, et au fur et à mesure des besoins, l'énergie qu'elle est susceptible de fournir par transmutations.

Pour réduire la vitesse de tout ou partie des neutrons émis, on introduit au sein de la masse d'uranium — et suivant une répartition qui n'est pas nécessairement uniforme — des éléments très légers tels que l'hydrogène, le deutérium par exemple, libres ou combinés, ou des éléments légers tels que le glucinium, le carbone, l'oxygène par exemple, libres ou combinés.

Un avantage spécial des ralentisseurs très légers est de diminuer la proportion des neutrons qui sont absorbés par résonance dans l'uranium et qui peuvent être ainsi perdus pour le processus en chaînes.

Ces éléments pourront être introduits sous forme liquide, gazeuse ou solide (poudre par exemple).

Ils peuvent être mélangés plus ou moins intimement avec l'uranium ou le composé d'uranium, et ce mélange peut être obtenu par tous procédés connus.

On peut, par exemple, mélanger avec un composé d'uranium en poudre un composé hydrogéné sous forme solide, liquide ou gazeuse; ce composé

***Figure 5:** First page of the invention patent of an "Energy production device", requested by F. Joliot and his team on 1st May 1939.*