

# FRANCE'S CONTRIBUTION TO THE DISCOVERY OF THE CHAIN REACTION

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One of the characteristics of modern science and technology is the extraordinary overlapping pattern of the contributions by different teams working on the same problem in different countries. As a result, it is difficult, and sometimes even presumptuous, to endeavour to remove from its international context and single out the contribution made to a common problem by one particular team. I shall nevertheless attempt here to describe the part played by French scientists in the series of developments that led to the epoch-making start-up of Fermi's reactor in Chicago on 2 December 1942, the event that was the genesis of the atomic problem, on the evolution of which the fate of our civilization depends.

The real beginnings of the atomic adventure go back to the discovery of radioactivity and radium by Henri Becquerel and Pierre and Marie Curie in France at the end of the last century. This was the crucial step - henceforth the way to the new alchemy was open and the key to the structure of matter lay within the reach of modern science, which was to progress in particular through the brilliant discoveries of Ernest Rutherford and Niels Bohr along the long road which we are still travelling today. The final, indispensable stage preceding the discovery of fission was completed at the Radium Institute in Paris in February 1934, a few months before the death of Marie Curie. Using the unrivalled stocks of radioactive materials accumulated with characteristic patience and tenacity over a period of more than 30 years by Madame Curie, the son-in-law and daughter of the scientist, Frédéric and Irène Joliot, prepared the most powerful source of polonium hitherto available; on exposing aluminium to that source they found that atoms were produced belonging to a radioactive isotope of phosphorus unknown in nature. They had just discovered artificial radioactivity, which was to make it possible for science to pass from the stage of natural to that of controlled alchemy.

In the address which he delivered on receipt of the Nobel prize in 1935, Joliot was already able to foresee the next stage: "If we look back at the progress that has been achieved by science, and achieved at an ever-increasing pace, we are justified in believing that scientists, breaking and making atoms at will, one day will succeed in bringing about explosive nuclear chain reactions. If it becomes possible for



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transmutations of this kind to be propagated in matter, one can imagine what enormous amounts of useful energy will be liberated."

Soon after Joliot's discovery in 1934, Enrico Fermi, in Rome, began to study the transmutations produced by neutron bombardment, and showed that in the case of uranium not one but twenty new radioactive substances were formed, the nature and origin of which could not be explained.

It took nearly five years to find the answer to this riddle, which was studied in Berlin by Otto Hahn and his co-workers, Lise Meitner and Fritz Strassmann. At the end of the first year these scientists believed that they had solved the chemical puzzle of identifying the new radioactive elements by classifying them as substances ranging from the last known positions in the periodic table to those of various unknown elements

with atomic numbers higher than that of uranium.

In the following year, sceptical about these results of Hahn, Irène Curie, assisted by the Yugoslav physicist Paul Savitch, concentrated her efforts on identifying just one of the radioelements in question, since it seemed to her to be impossible to deal with the whole chemical complex at once. The problem was a difficult one and Irène Curie twice put forward solutions which were later disproved; this did not discourage her, however, and with a persistence and a patience worthy of her mother she finally succeeded in 1938 in proving that the chemical properties of the radioelement under consideration were very similar to those of a known element, lanthanum, situated in the middle of the periodic system. This hypothesis was completely at variance with the theory advanced earlier by the Berlin school.

Although they were convinced that Irène Curie's results were wrong, Hahn and Strassmann nevertheless felt obliged in the light of her work to re-appraise their earlier conclusions; this was to put them eventually on to the right track. Using the same process of fractional crystallization which forty years earlier had enabled Pierre and Marie Curie to isolate radium, Hahn showed that the chemical behaviour of the radioelements formed by the action of neutrons on uranium was identical to that of elements situated in the middle of the periodic table. One month after the publication of Hahn's results at the end of 1938, i. e. in January 1939, physical evidence for the break-up of the uranium nucleus was provided by Otto Frisch in Copenhagen, a fortnight later by Frédéric Joliot working independently in Paris, and then by other scientists in the United States and England.

This was followed a month later by the discovery at the Collège de France by Joliot and his two co-workers, Hans Halban and Lew Kowarski, of a new phenomenon of fundamental importance, viz. that the fission of the uranium nucleus is accompanied by the emission of secondary neutrons.

Overnight, atomic physics ceased to be the realm of basic research and the prerogative of the isolated research worker. A new élite, that of nuclear scientists imbued with an awareness of their moral and political responsibilities, was about to be born and play a role of increasing importance in the lives of the great nations.

A new era had dawned in which, in certain fields of research, progress was to necessitate the mobilization of the entire resources of a country or even of several countries; atomic energy was to be the first example of this development.

Starting in the spring of 1939, scientists in the more advanced countries began to warn their governments, making every effort to convince them of the importance of uranium in the civilian and the military

sphere and seeking to obtain the assistance they needed in order to continue their investigations on a larger scale.

The man most obsessed at the time by the potential power of a nuclear weapon was the Hungarian physicist Leo Szilard. In February 1939 he made contact, from New York, with scientists living in the countries which were later to become allies in the war which he considered inevitable. He suggested that they agree to cease publication of all material dealing with nuclear fission. I well remember the surprise that was caused at Joliot's laboratory at the Collège de France when a telegram of over 140 words - the longest we had ever seen - arrived from a colleague of Szilard, and the discussion that followed on whether or not it would be possible to obtain voluntary general agreement on the maintenance of secrecy with regard to current and future research. The idea did not seem feasible in nuclear physics, which to that date had been the domain of pure science par excellence. The exchange of information had always been entirely unrestricted and at times had even taken on the character of a race, in which a few days earlier or later in dispatching a letter to a scientific periodical might mean, for the writer, the difference between the fame of making a discovery himself and the lesser satisfaction of merely confirming it. This is what in fact had just happened with the discovery of secondary neutrons, which was announced by the Joliot team in Paris on 3 March 1939 and by Szilard and Fermi in New York a week later.

Szilard's proposal was not fully understood or accepted, but a few months later, just before the war, each country began independently to treat the results of its uranium research as secret information.

The French research work went on throughout the summer of 1939, Joliot's team being joined by Francis Perrin, who was the first to enunciate the principle of the critical mass required for the chain reaction. This team worked on determining experimentally how many secondary neutrons were emitted per fission and arrived at an estimate of 3 to 4 which, however, subsequently proved to be too high. It was soon concluded from these results that if a chain reaction was to be produced in natural uranium it would be necessary to mix the uranium with some substance composed of light elements able to slow down the neutrons without absorbing them excessively. This principle formed the basis of the first patent applications, filed in France at the beginning of May 1939; one of them dealt with the use of uranium as an explosive, while the others concerned uranium-based power-producing devices, later known as atomic piles or reactors. These patents, the first of their kind to be filed anywhere in the world, are recognized by numerous countries. The property of the French nation, they foretold the subsequent development

of the nuclear weapons and nuclear installations now in existence.

In the autumn of 1939 came the war, and this gave an added impetus to research activities. There was no question in France at this time of seeking to make a bomb, which was regarded as too difficult, but consideration was given to the possibility of constructing power generators: the technicians were not fully aware of the difficulties of the problem and they believed that a submarine propulsion unit, which would have the enormous advantage of not requiring an oxygen supply, might be feasible within a few years.

The first substance envisaged for use as a moderator was hydrogen but an investigation of ordinary water-uranium mixtures soon showed hydrogen to be too avid an absorber of neutrons for the purpose. Finally, Joliot's team came to the conclusion that the best moderator would be deuterium oxide, the "heavy water" discovered by the American physicist Harold Urey in 1932. On the eve of the war this substance cost half a dollar per gram and was only used for research purposes. Nevertheless, even with no prospective market, a Franco-Norwegian synthetic ammonia company had gone ahead with the manufacture of heavy water on a kilogram scale, making use of the low cost of electricity in Norway and a system of fractional electrolysis associated with the main activity, ammonia production.

The French Ministry of Munitions was informed of the research in progress and with its support a secret mission left for Oslo in March 1940, a few weeks before the invasion of Norway, to bring back 165 liters of heavy water, the only stock of this valuable substance then in existence.

The French Government was fully alive to the importance of the problem and gave Joliot exceptional facilities, including unlimited funds and release from military service of any co-worker whom he needed. It also initiated scientific collaboration with Great Britain by sending a representative to London for a few hours in April 1940 to report on the first French results. At the same time, contracts were concluded with the Norwegian firm reserving for France the entire production of heavy water in the years ahead; another contract, negotiated with Belgian industry, would have secured the uranium production of the Belgian Congo, the richest source of uranium in the world at the time. Independently of this, six tons of Belgian uranium oxide were delivered to Joliot.

Thus, both in the technical field and in that of raw material supplies, the French effort appeared to have got off to an excellent start. Unfortunately, its development was then halted by the invasion of the country.

Joliot's laboratory and the heavy water were first withdrawn to Clermont-Ferrand and then, on 16 June 1940, a grave decision was taken at Bordeaux:

Halban and Kowarski were to leave for England taking the precious cargo with them; there they were to put themselves at the disposal of the British authorities and continue the experiments begun in France. Joliot, who did not fully realize the vital role he could have played in the work in the United Kingdom and the United States, decided to remain in charge of his laboratory in occupied France.

In Britain there was an equally keen awareness of the importance of the uranium problem, but the MAUD Committee set up in April 1940 to study the question had devoted more attention to the military possibilities than to energy production. Halban and Kowarski, together with their heavy water and their plans, were warmly welcomed and a laboratory was placed at their disposal at Cambridge. It was there in 1940 that they performed the experiment that had been planned with Joliot. Thanks to a neutron study of a uranium oxide suspension in an aluminium sphere filled with heavy water, they were the first, if not actually to produce experimentally a critical, chain-reacting pile, at least to show almost conclusively that such a reaction was possible in a natural uranium and heavy water system but that the critical mass for such a system would necessitate tons of each component. This was a far cry from the 165 liters of heavy water actually available.

Considerable importance attaches to this experiment, the results of which were used on a number of occasions by the American scientists as an argument to persuade their government in its turn to embark on a large-scale project.

As early as October 1939, President Roosevelt had been alerted by a letter from Einstein and a report from Szilard; the latter referred to the French work as being probably the most advanced at the time.

When the results of the Halban-Kowarski experiment became known in the United States in the spring of 1941, the question arose as to whether it would not be preferable to explore the heavy water rather than the graphite approach, which was being studied by Fermi and Szilard, as a means of achieving a self-sustaining chain reaction.

As it was impossible to manufacture heavy water on an industrial scale in 10-ton quantity in less than nearly three years, the advocates of graphite won priority over those of heavy water, the principal American champion of which was Urey himself, who, like Halban, feared that neutron absorption by the graphite would be too high to enable a self-sustaining chain reaction to take place with natural uranium. During the whole of the period from 1941 to the middle of 1942, when it was still impossible to tell with certainty from Fermi's graphite-uranium exponential experiments at Columbia University whether criticality was possible with this system, heavy water remained as a stand-by solution for the team working on the possibilities of plutonium production.

Having been seconded by the Free French Forces to the British atomic group, I was fortunate enough to be sent by the latter to the Metallurgical Project in Chicago in July 1942. Over 100 scientists were already at work in the various laboratories of the University and throughout this young and enthusiastic group the atmosphere was excellent.

The plans at this time, which were to be implemented with miraculous precision, provided for the production of the first few kilograms of pure fissile material within three years. The engineers knew that they were engaged on the development of explosives which would have an unprecedented power of destruction, but their moral scruples were stifled partly by the fear - which subsequently proved unfounded - that Germany was working along similar lines and might even be in the lead, and partly by the immense fascination of the work itself.

We often used to go down to the large shed erected beneath the stands of the University football ground, where the uranium-graphite structure - later given the name of atomic pile by Fermi - was being assembled in the greatest secrecy.

In a mysterious enclosure gleaming with graphite powder, a group of men, black from head to foot, were working under conditions of strict secrecy on the erection of a shining black structure several metres square made up of graphite bricks, some of which were hollow and contained uranium metal or compressed uranium oxide. The strange construction was a moving sight since we all knew that on it there perhaps depended the outcome of the war and with it the future of mankind.

Elsewhere, the Seaborg team (to which I had been assigned and which comprised about 30 chemists of whom the eldest, its chief, was aged 30) was succeeding in isolating one-fifth of a milligram of plutonium, the first ever to be visible to the naked eye, from several hundred kilograms of uranium salts that had been bombarded by the California cyclotron, the most powerful in the world at the time. When on 18 August 1942, at the weekly meeting of the graduate scientists engaged on the project (attended by a number of workers which increased from session to session at a rate worthy of the chain reaction itself), Seaborg rose to announce that he had for the first time seen a minute quantity of a pinkish-coloured plutonium salt, the product of a man-made transmutation, Edward Teller, the head of the theoretical physics group, asked what particular salt it was; Seaborg replied that he

was not allowed to say - an example of the strict compartmentalization of information practised as an anti-leakage measure.

During this same summer of 1942 the American advocates of the heavy water pile had continued their efforts and had succeeded in having a number of decisions taken regarding the industrial production of this type of moderator in Canada and then in the United States. At one time there was a plan to put an English team under Halban to work within the Metallurgical Project in Chicago, but this scheme was abandoned by the British and replaced by a decision to launch a major project in Canada, which would be able to benefit from the proximity of United States research centres and the facilities available in North America but no longer possible in a United Kingdom fully mobilized for war.

This was the beginning of the Anglo-Canadian project which, originally set up in Montreal in November 1942, led two years later to the establishment at Chalk River of a great Canadian nuclear research centre devoted exclusively to heavy water reactors and power plants. Up to the middle of 1944 the director of the project was Halban himself, who was then succeeded by Sir John Cockcroft. The team included four other French members: Pierre Auger, in charge of physics; Lew Kowarski, who led the group responsible for construction of the first Canadian heavy water pile, ZEEP, commissioned in September 1945, being also the first pile to operate outside the United States (the first heavy water pile in the world, CP-3, went critical in Chicago in May 1944); and two chemists, Jules Guéron and myself; I was responsible for developing in Canada some of the earliest techniques for organic solvent extraction of plutonium. All four of us returned to France in 1946 to take part in the establishment of the Commissariat à l'Énergie Atomique, whose first achievement was the bringing into operation in December 1948 of a zero-power heavy water pile.

Thus, the work of French scientists, while it did not contribute directly to the brilliant success of 2 December 1942, did nevertheless play an important role: on the one hand, it served to back up the efforts of Great Britain, which in turn were partly instrumental in persuading the United States to tackle the uranium problem on an industrial scale; and on the other hand, it played a large part in the birth of nuclear activities in Canada and in the development of heavy water reactors.