# INITIATION OF THE CHAIN REACTION

## The Search for Pure Materials

by

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During the past twenty years many accounts of the events which led to the first successful chain reaction have appeared. The first of these was the famous Smyth Report, in 1945, but since then Professor A. H. Compton's book "Atomic Quest" and General Groves' "Now It Can Be Told" have become available. "The New World" by R. G. Hewlett and O. E. Anderson Jr., written under the auspices of the Historical Advisory Committee of the US Atomic Energy Commission, has appeared and is an exhaustive and scholarly treatment by trained historians who had access to thousands of pertinent documents. In the face of these carefully prepared statements, it is futile to write, in a short article such as this, a summary with claims to scholarly accuracy.

It is widely known that the first sustained chain reaction took place under war-time secrecy in an aggregate of graphite, uranium metal and uranium oxide. The construction of the "pile" was part of the programme of the so-called Metallurgical Project, which was eventually sponsored by the US Army Corps of Engineers. An outstanding group of physical scientists, including Enrico Fermi, had been gathered and was given almost unlimited financial support. The following lines are in no sense an adequate summary of their activities prior to 2 December 1942; on the contrary, they are casual observations by one participant, including some very minor details which - I hope - will give some personal interest to the otherwise factual account.

My first important assignment under the programme was to repeat, with minor variations, some of the work of Fermi and his group at Columbia. We planned to use the old Chicago cyclotron as a neutron source and study neutron diffusion in graphite with and without a uranium-bearing lattice embedded in it. Wiser heads than ours knew that with a cyclotron built in 1937, as ours was, most of our time would be spent in maintenance, and little or none on diffusion measurements. More or less unexpectedly, certainly without any urging from me, a gram of radium, in the form of a mixed neutron source, arrived from Washington and we released the cyclotron for other projects.

I made several trips to New York to keep in touch with Fermi's group. They had assembled and tested a sub-critical "exponential" pile of uranium

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oxide powder packed in aluminium boxes stacked in a graphite matrix. The neutron reproduction factor "k", which had to exceed unity for success, was 0.88. Fermi considered this as not very encouraging but was going ahead with the same materials, with certain improvements. In particular, he was dissatisfied with the quality of the oxide, and later events showed his suspicions to have been correct. Furthermore, there were calculable advantages to be gained from compressing the oxide powder to high density, and Anderson and Zinn were looking out for a used press for this purpose. One benefit of pressing into briquets would be to get rid of the boxes, which absorbed some of the precious neutrons. The advantages of high density powder briquets were so obvious that I decided to make pressings for the first pile at Chicago.

The uranium oxide powder used at Columbia and Chicago came from Canadian pitchblende, and was es-

sentially the residuum of a separation in which all emphasis had been put on recovering the radium. Fermi asked some of the Columbia chemists to analyse it, and the list of impurities was so long that it looked like a fair sample of the periodic system. The original shipment of oxide to Columbia had, however, one favourable property; it pressed beautifully into nice stable briquets. At Chicago, working with a batch of Canadian oxide shipped about a year later, the pressing programme ran into trouble. The briquets were not nearly so stable as their predecessors had been at Columbia, and often broke at the slightest touch. They were so delicate that we were forced to take a step backward and enclose them in thin-walled aluminium boxes, held together with Scotch tape, at that time a relatively new product. The pressing difficulties at Chicago elicited some good humoured but uncomplimentary remarks from Columbia, but a serious effort to find the cause by comparison of the pressing procedures failed to give any explanation.

At a time when the pressing of the oxide at Chicago was almost complete I stopped at Princeton on one of my eastern trips, and was promptly and vigorously set upon by Wigner and Creutz. Wigner was sure that Fermi's graphite-to-uranium ratio at Columbia was off the optimum, and in particular that Fermi's oxide briquets were too large. I had brought along one of the Chicago briquets, and when it was displayed Wigner said it was too big and should be cut into two. I launched into a tear-jerking story of the difficulties of the Chicago pressing, how the lumps would just barely cohere, and how they would disintegrate to powder if sawing were tried. During this monologue I did not notice that Creutz had quietly disappeared, taking my sample briquet with him. In a few minutes he brought it back, sawed neatly into two, with both halves intact. I went home and we cut all our precious pressings to the Wigner prescription.

When the first Chicago exponential pile was assembled, and the attenuation of the neutron flux measured by placing indium foils along its axis in the prescribed manner, "k" seemed to be about 0.91. Fermi came from Columbia to give the official approval, and asked for all the original data on the foils, their thicknesses, their positions, and their counting rates, which, of course, we gladly furnished him. He retired to an office and in a few hours called some of us in for a little seminar. After some preliminaries he announced that "k" was 0.93. In some surprise, I demurred and said it was 0.91. On only one other occasion did I ever see Fermi so annoyed. He always prided himself on being more conservative than anyone else, and here was a situation in which an experimenter was more cautious about his own result than was Fermi in his evaluation of it. After some sharp remarks he began to smile again, and we found that I had been using an older value of the graphite diffusion coefficient, which Fermi had abandoned.

At first the news of the higher "k" at Chicago had a depressing effect at Columbia, as if some technical break-through had bypassed them, the originators of the programme. The explanation was soon at hand in that the Canadians had been doing some refining of oxide between the Columbia and the Chicago shipments. Although impure by modern standards the Chicago oxide was much better than the Columbia; in particular, it lacked a kind of tarry impurity that explained the superior pressing qualities of the Columbia material.

But by now it had been decided to concentrate the entire effort on the chain reaction at Chicago, and soon Fermi and his group arrived from Columbia. They were by now professionals in the art of the exponential pile, at which I was only an apprentice, and I left the graphite stacking to the newcomers. It was becoming more and more apparent that the critical difficulty was to obtain, in sufficient quantity, high purity uranium compounds and uranium, and graphite. Our early attempts to produce pure uranium metal were not very successful. Due to its high efficiency for generation of X-rays from electron bombardment, the Westinghouse Lamp Works had made gram amounts of it for testing as X-ray tube targets. Unfortunately for this application it was found that the melting point of the pure metal was at least 600°C lower than reported in the literature. The Westinghouse process involved a photochemical reduction of uranyl nitrate in a solution containing potassium fluoride and sugar. The $KUF_5$ which precipitated out was fused and uranium recovered by electrolysis of the molten salt. The resulting metal was the best available in the early stages of the project, but the process was too difficult to expand to produce the desired quantities.

A method in which uranium oxide was reduced to powdered uranium by heating with lithium hydride, and the powder subsequently sintered into lumps of metal, produced very poor material, some of which was pyrophoric when the containers were opened to air. It was not sufficient to find a process for producing uranium, it was also necessary to evolve and use analytical techniques sufficiently sensitive to control the purity to the high specifications. The old analytical methods for boron were inadequate, and new ones were developed at the Bureau of Standards. Methods for cadmium were tested at Princeton.

I resolved to devote myself to the problem of pure materials, and this seemed logical, for at this time Professor Compton asked me to be chairman of the chemistry section of the rapidly expanding project. The haste and confusion were incredible. We had learned from England a method of producing uranium by the reduction of the tetrafluoride with calcium or magnesium. Dr. Alexander Smith came out of retirement and set up for us a small pilot plant for producing the fluoride from the nitrate. A great leap

forward occurred when Compton persuaded the Mallinckrodt Chemical Company to agree to purify uranyl nitrate by the hazardous method of ether extraction. Contracts were let to various companies for uranium production, but methods of analysis were at first not circulated among them. For some months I published a weekly analytical review, recording the results of analyses of uranium preparations made at Chicago, the Bureau of Standards, Princeton, St. Louis, Ames, and many other places where analysis and production were under way. Some of the companies did not like having their analytical procedures made available to their competitors, but the urgency of the development overwhelmed all objections.

The purity of the graphite was of equal importance. It was very difficult to explain to the big graphite producers whose product had been perfectly satisfactory for electrodes in the steel industry why thousands of tons had to be prepared to the standards of graphite for arc terminals in spectroscopic analysis. Nobody had ever worried about boron in graphite before; nobody knew how much boron commercial graphite contained. I remember an exhausting trip with Norman Hilberry to the Speer Graphite Company, located in an obscure region of northwest Pennsylvania. This rather small concern had agreed to test some procedures we recommended in the production of graphite to lower the boron content. I remember the excitement when the first shipment of Speer graphite arrived in May 1942 and "k" soared to 0.995, still using sub-standard uranium.

I am, of course, only tracing a thread through the rapidly proliferating project. The chemists were separating plutonium in microgram amounts from material irradiated at the Washington University cyclotron. Others were finding new fission products daily, or studying the dislocation effects in graphite caused by fast neutron bombardment. Physicists looked for and found new delayed neutron emitters, and measured the decay curve of the sum of all fission products. Lattices with variable uranium-to-graphite ratios were built to test the effect on "k". Biologists were beginning tests of the health hazards from slow neutrons in order to design the reactor shield. Reports of an extended conference at Berkeley where the possibility of the thermonuclear reaction had been discussed seeped through the laboratory in spite of being officially withheld. Vigorous debates on liquid versus gas cooling of reactors were a daily occurrence.

Looked at in retrospect, the programme of the Metallurgical Project seems to have been efficiently streamlined to attain its goal, but to one enmeshed in the turmoil at that time there seemed long periods of no progress, and to me at least there was a sense of almost unbearable confusion, urgency, and fear that our opponents in the war were far ahead of us. At last, on 2 December 1942, enough graphite, uranium and uranium oxide of sufficient purity had arrived at Chicago so that when the most recent shipment was added to the growing structure of the reactor, and Fermi had the control rod pulled out, the neutron flux rose exponentially with the positive second time derivative that Wigner insisted on seeing. But there were many conferences going on that day and few could afford the time to be present. In another room the duPont reviewing committee was in session and hearing statements from project members. John Marshall, for instance, who had worked as hard as anyone on the actual pile was temporarily away at the Metal Hydrides plant, and plans for reactors were being debated in many offices.

On that day I came home late as usual, exhausted as usual, and since I was not permitted to say a word about our work to my wife and family, fell into bed to gather strength to face the chaos to be expected, as usual, on the following morning.