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Atomic Energy for Military Purposes

The Official Report
on the Development of the Atomic Bomb
under the Auspices
of the United States Government,
1940–1945

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CHAPTER VI. THE METALLURGICAL PROJECT AT CHICAGO IN 1942

INTRODUCTION

6.1. As has been made clear in Chapters IV and V, the information accumulated by the end of 1941 as to the possibility of producing an atomic bomb was such as to warrant expansion of the work, and this expansion called for an administrative reorganization. It was generally accepted that there was a very high probability that an atomic bomb of enormous destructive power could be made, either from concentrated U-235 or from the new element plutonium. It was proposed, therefore, to institute an intensive experimental and theoretical program including work both on isotope separation and on the chain-reaction problems. It was hoped that this program would establish definitely whether or not U-235 could be separated in significant quantities from U-238, either by electromagnetic or statistical methods; whether or not a chain reaction could be established with natural uranium or its compounds and could be made to yield relatively large quantities of plutonium; and whether or not the plutonium so produced could be separated from the parent material, uranium. It was hoped also that the program would provide the theoretical and experimental data required for the design of a fast-neutron chain-reacting bomb.

6.2. As has been explained in Chapter V, the problems of isotope separation had been assigned to groups under Lawrence and Urey while the remaining problems were assigned to Compton's group, which was organized under the cryptically named "Metallurgical Laboratory" of the University of Chicago. In this chapter and the following two chapters we shall describe the work of the Metallurgical Laboratory and the associated laboratories up to June 1945. In later chapters we shall discuss isotope-

separation work and the work of the bomb development group, which was separated from the Metallurgical Laboratory early in 1943.

6.3. It would be futile to attempt an assessment of the relative importance of the contributions of the various laboratories to the overall success of the atomic-bomb project. This report makes no such attempt, and there is little correlation between the space devoted to the work of a given group and the ability or importance of that group. In deciding which subdivision of the atomic-bomb project should be discussed first and most fully, we have been governed by criteria of general interest and of military security. Some developments of great technical importance are of little general interest; others both interesting and important must still be kept secret. Such criteria, applied to the objectives and accomplishments of the various laboratories set up since large-scale work began, favor the Metallurgical Laboratory as the part of the project to be treated most completely.

OBJECTIVES

6.4. In accordance with the general objectives just outlined, the initial objectives of the Metallurgical Laboratory were: first, to find a system using normal uranium in which a chain reaction would occur; second, to show that, if such a chain reaction did occur, it would be possible to separate plutonium chemically from the other material; and, finally, to obtain the theoretical and experimental data for effecting an explosive chain reaction with either U-235 or with plutonium. The ultimate objective of the laboratory was to prepare plans for the large-scale production of plutonium and for its use in bombs.

ORGANIZATION OF THE WORK

6.5. The laboratory had not only to concern itself with its immediate objectives but simultaneously to bear in mind the ultimate objectives and to work toward them on the assumption that the immediate objectives would be attained. It could not wait for a chain reaction to be achieved before studying the

chemistry of plutonium. It had to assume that plutonium would be separated and to go ahead with the formulation of plans for its production and use. Consequently problems were continually redefined as new information became available, and research programs were reassessed almost from week to week. In a general way the experimental nuclear physics group under E. Fermi was primarily concerned with getting a chain reaction going, the chemistry division organized by F. H. Spedding (later in turn under S. K. Allison, J. Franck, W. C. Johnson, and T. Hogness) with the chemistry of plutonium and with separation methods, and the theoretical group under E. Wigner with designing production piles. However, the problems were intertwined and the various scientific and technical aspects of the fission process were studied in whatever group seemed best equipped for the particular task. In March 1942, Thomas Moore was brought in to head the engineering group. Other senior men in this group were M. C. Leverett, J. A. Wheeler and C. M. Cooper, who later succeeded Moore as head of the Technical Division. In the summer of 1942 the importance of health problems became apparent and a Health Division was organized under Dr. R. S. Stone. The difficult task of organizing and administering a research laboratory growing in size and complexity with almost explosive violence was carried out by R. L. Doan as Laboratory Director.

6.6. We have chosen to confine this chapter to the work of 1942 because a self-sustaining chain reaction was first achieved on December 2 of that year, at a time when the whole Chicago project was being appraised by a reviewing committee with the members particularly selected for their engineering background.* That was a dramatic coincidence and also a convenient one for purposes of this report since either incident might be considered to mark the end of an epoch at the Metallurgical Laboratory. Furthermore, in preparation for the reviewing committee's visit

* This committee was composed of W. K. Lewis, C. H. Greenewalt, T. C. Gary, and Roger Williams. E. V. Murphree was also a member but due to illness was unable to participate.

a comprehensive report had been prepared. That report was generally known as the "Feasibility Report" and has been used extensively in preparing this chapter.

PLAN OF THIS CHAPTER

6.7. In this chapter we shall present the material in the order of the objectives given above. In Part I we shall discuss progress towards the initial objectives, including (a) procurement of materials, (b) the experimental proof of the chain reaction, (c) the chemistry of plutonium and some of the problems of separation, (d) some of the types of auxiliary experiments that were performed, and finally (e) the "fast neutron" work. Necessarily the work described in detail is only a sampling of the large amount of theoretical and experimental work actually performed. In Part II we shall discuss the possibilities that were considered for production piles and separation methods, and the specific proposals made in November 1942.

PART I: PROGRESS TOWARD THE INITIAL OBJECTIVES

PROCUREMENT OF MATERIALS

General

6.8. It has been made clear in earlier chapters of this report that the procurement of materials of sufficient purity was a major part of the problem. As far as uranium was concerned, it seemed likely that it would be needed in highly purified metallic form or at least as highly purified uranium oxide. The other materials which were going to be needed were either graphite, heavy water, or possibly beryllium. It was clear at this time that, however advantageous heavy water might be as a moderator, no large quantities of it would be available for months or years. Beryllium seemed less advantageous and almost as difficult to get. Therefore the procurement efforts for a moderator were centered on graphite. As has been explained in Chapter V, procurement of

uranium and graphite was not primarily the responsibility of the Metallurgical Laboratory but was handled through E. V. Murphree and others on the "planning board." In fact, the obvious interest of the Metallurgical Laboratory in the problem led to continual intervention by its representatives. A great deal of the credit for the eventual success in obtaining materials is due to N. Hilberry and later R. L. Doan, always supported by A. H. Compton.

Uranium Ore

6.9. Obviously there would be no point in undertaking this whole project if it were not going to be possible to find enough uranium for producing the bombs. Early indications were favorable, and a careful survey made in November 1942 showed that immediate delivery could be made of adequate tonnages of uranium ores.

Uranium Oxide and Uranium Metal

6.10. At the end of 1941 the only uranium metal in existence was a few grams of good material made on an experimental basis by the Westinghouse Electric and Manufacturing Company and others and a few pounds of highly impure pyrophoric powder made by Metal Hydrides Company. The only considerable amount of raw material then available in this country was in the form of a commercial grade of black uranium oxide, which could be obtained in limited quantities from the Canadian Radium and Uranium Co. It contained 2 to 5 per cent of impurities and was the material which gave a neutron multiplication factor of only about 0.87 when used in an exponential pile.

6.11. By May 1942, deliveries averaging 15 tons a month of black oxide of higher purity and more uniform grade started coming in. Total impurities were less than 1 per cent, boron comprised a few parts per million, and the neutron multiplication factor (k) was about 0.98. (It is to be remembered that the multiplication factor depends also on the purity of the graphite.) Deliveries of this material reached a ton a day in September 1942.

6.12. Experiments at the National Bureau of Standards by J. I. Hoffman demonstrated that, by the use of an ether extraction method, all the impurities are removed by a single extraction of uranyl nitrate. The use of this method removed the great bulk of the difficulties in securing pure oxide and pure materials for the production of metal. Early in May 1942, arrangements were completed with the Mallinckrodt Chemical Works in St. Louis to put the new grade of oxide through an ether extraction process on a production basis for a further reduction in impurity content and to deliver the final product as brown dioxide. Deliveries started in July 1942 at a rate of 30 tons a month. This oxide is now used as a starting point for all metal production, and no higher degree of purity can be expected on a commercial scale. In fact, it was a remarkable achievement to have developed and put into production on a scale of the order of one ton per day a process for transforming grossly impure commercial oxide to oxide of a degree of purity seldom achieved even on a laboratory scale.

6.13. The process which Westinghouse had been using to produce the metal was the electrolysis of KUF_5 at a cost of about $1,000 a pound. Since the KUF_5 was produced photochemically under the action of sunlight this method constituted a potential bottleneck in production. It was found that uranium tetrafluoride could be used instead of KUF_5, and steps were taken to have this salt produced at the Harshaw Chemical Company in Cleveland and at the du Pont plant in Penns Grove, New Jersey. Production started in August 1942 and by October 1942 was up to 700 pounds per day at Harshaw and 300 pounds per day at du Pont, the method of manufacture in both cases being the hydrofluorination of Mallinckrodt-purified dioxide.

6.14. As the result of this supply of raw materials to Westinghouse, and as a result of plant expansion, deliveries from Westinghouse had accumulated to a total of more than 6,000 pounds by November 1942 and were expected to be at the rate of 500 pounds per day by January 1943. The purity of the metal was good, and the cost had dropped to $22 per pound.

6.15. Deliveries of acceptable metal from Metal Hydrides Co. were delayed for various reasons and were just beginning in November 1942. This company's production was supposed to reach a thousand pounds per week thereafter.

6.16. Neither the Westinghouse process nor the Metal Hydrides Process was entirely satisfactory. Intensive activity designed to accelerate metal production, and carried out independently by F. H. Spedding and his associates at Iowa State College at Ames, Iowa, and by C. J. Rodden at the National Bureau of Standards, resulted in the development of a satisfactory method. Production facilities were set up at Ames in the fall of 1942 and had already produced more than one ton by the end of November. The process was extremely simple, rapid and low cost.

6.17. Further research indicated additional changes that could be made to advantage, and by the middle of 1943 Spedding at Iowa and other producers who entered the picture were using the final production method adopted.

6.18. By the end of 1942 arrangements had been made by the Manhattan District to increase metal production by making greater use of the Mallinckrodt Chemical Works, the Union Carbide and Carbon Corporation, and the du Pont Company.

6.19. To summarize, almost no metal was available during most of 1942, a fact that seriously delayed progress as we shall see, but the production problems had been nearly solved by the end of 1942 and some 6 tons of metal were incorporated in the pile built in November 1942. The whole problem of procurement of metal was taken over by the Manhattan District at the end of the year, under the general direction of Colonel Ruhoff, formerly with the Mallinckrodt Chemical Works. From the point of view of the Metallurgical Project no further serious delays or difficulty have occurred because of metal shortages.

Graphite Procurement

6.20. At the beginning of 1942 graphite production was still unsatisfactory but it was, of course, in quite a different condition from the metal production since the industrial production of

graphite had already been very large. The problem was merely one of purity and priority. Largely through the efforts of N. Hilberry, the National Carbon Company and the Speer Carbon Company were both drawn into the picture. Following suggestions made by the experts of the National Bureau of Standards, these companies were able to produce highly purified graphite with a neutron absorption some 20 per cent less than the standard commercial materials previously used. Although efforts further to reduce the impurities have had some success, the purity problem was essentially solved by the middle of 1942 and large orders were placed with the cooperation of the War Production Board. As in the case of the metal, the graphite procurement problem was taken over by the Manhattan District.

THE CHAIN REACTION

Further Intermediate Experiments

6.21. At the time that the Metallurgical Project was organized, most of the physicists familiar with the problem believed that a chain-reacting pile probably could be built if sufficiently pure graphite and pure uranium metal could be obtained. Enough work had been done on resonance absorption, on the theory of absorption and diffusion of neutrons in a pile, and on intermediate experiments to make it possible to design a lattice structure that had a very good chance of maintaining a chain reaction. Nevertheless, there were uncertainties in the experimental data and in the approximations that had to be made in the theoretical calculations. There were two alternatives: (1) to build a pile according to the best possible design; (2) to make more accurate determinations of the pertinent nuclear constants, to perform intermediate experiments, and to improve the calculations. There is little doubt that the first alternative was the one likely to lead most rapidly to the production of plutonium. There were many important questions which could have been answered more rapidly by such an operating pile than by a series of small-scale experiments. Unfortunately, the necessary amounts of materials

were not available and did not become available for nearly nine months. Consequently, it was necessary to choose the second alternative, that is, to accumulate all relevant or possibly relevant information by whatever means were available.

6.22. The major line of investigation was a series of intermediate experiments. The particular set-up for each intermediate experiment could be used to test calculations based on separate auxiliary experiments. For example, the proportion of uranium oxide to graphite was varied, oxides of different purities were used, oxide was used in lumps of various sizes and shapes and degrees of compression, the lattice spacing was varied, the effect of surrounding the uranium oxide units with beryllium and with paraffin was tried; and, finally, piles of identical lattice type but of different total size were tried to see whether the values of the multiplication factor k (for infinite size) calculated from the different sets of results were identical. In general, E. Fermi had direct charge of investigations of effects of impurities, and S. K. Allison had charge of tests involving different lattice dimensions. All these experiments strengthened the confidence of the group in the calculated value of k and in the belief that a pile could be built with k greater than unity. In July enough purified uranium oxide from Mallinckrodt was available to permit building intermediate pile No. 9. As in previous experiments, a radium-beryllium neutron source was placed at the bottom of the lattice structure and the neutron density measured along the vertical axis of the pile. By this time it was known that the neutron density decreased exponentially with increasing distance from the neutron source (hence the name often used for experiments of this type, "exponential pile") and that, from such rates of decrease, the multiplication constant k for an infinitely large pile of the same lattice proportions could be calculated. For the first time the multiplication constant k so calculated from experimental results came out greater than one. (The actual value was 1.007.) Even before this experiment Compton predicted in his report of July 1 that a k value somewhere between 1.04 and 1.05 could be obtained in a pile containing highly purified uranium oxide and

graphite, provided that the air was removed from the pile to avoid neutron absorption by nitrogen.

An Auxiliary Experiment; Delayed Neutrons

6.23. We shall not mention a majority of the various auxiliary experiments done during this period. There was one, however,—the study of delayed neutrons—that we shall discuss because it is a good example of the kind of experiment that had to be performed and because it concerned one effect, not heretofore mentioned, that is of great importance in controlling a chain-reacting pile.

6.24. From previous investigations, some of which were already published, it was known that about 1 per cent of the neutrons emitted in fission processes were not ejected immediately but were given off in decreasing quantity over a period of time, a fact reminiscent of the emission of beta rays from shortlived radioactive substances. Several half-lives had been observed, the longest being of the order of a minute.

6.25. It was realized early that this time delay gave a sort of inertia to the chain reaction that should greatly facilitate control. If the effective multiplication factor of a pile became slightly greater than 1, the neutron density would not rise to harmfully large values almost instantly but would rise gradually so that there would be a chance for controls to operate. (Other time intervals involved, such as those between collisions, are too small to be useful.)

6.26. Because of the importance of this effect of delayed neutrons for control it was decided to repeat and improve the earlier measurements. (The fact that this was a repetition rather than a new measurement is also typical of much of the work in physics at this period.) A description of the experiment is given in Appendix 3. The results indicated that 1.0 per cent of the neutrons emitted in uranium fission are delayed by at least 0.01 second and that about 0.7 per cent are delayed by as much as a minute. By designing a pile such that the effective value of k, the multi-

plication factor, is only 1.01 the number of delayed neutrons is sufficient to allow easy control.

The First Self-Sustaining Chain-Reacting Pile

6.27. By the fall of 1942 enough graphite, uranium oxide, and uranium metal were available at Chicago to justify an attempt to build an actual self-sustaining chain-reacting pile. But the amount of metal available was small—only about 6 tons—and other materials were none too plentiful and of varying quality. These conditions rather than optimum efficiency controlled the design.

6.28. The pile was constructed on the lattice principle with graphite as a moderator and lumps of metal or oxide as the reacting units regularly spaced through the graphite to form the lattice. Instruments situated at various points in the pile or near it indicated the neutron intensity, and movable strips of absorbing material served as controls. (For a more complete description of the pile, see Appendix 4.) Since there were bound to be some neutrons present from spontaneous fission or other sources, it was anticipated that the reaction would start as soon as the structure had reached critical size if the control strips were not set in "retard" position. Consequently, the control strips were placed in a suitable "retard" position from the start and the neutron intensity was measured frequently. This was fortunate since the approach to critical condition was found to occur at an earlier stage of assembly than had been anticipated.

6.29. The pile was first operated as a self-sustaining system on December 2, 1942. So far as we know, this was the first time that human beings ever initiated a self-maintaining nuclear chain reaction. Initially the pile was operated at a power level of ½ watt, but on December 12 the power level was raised to 200 watts.

Energy Developed by the Pile

6.30. In these experiments no direct measurements of energy release were made. The number of neutrons per second emitted

by the pile was estimated in terms of the activity of standardized indium foils. Then, from a knowledge of the number of neutrons produced per fission, the resultant rate of energy release (wattage) was calculated.

Conclusion

6.31. Evidently this experiment, performed on December 2 just as a reviewing committee was appraising the Chicago project, answered beyond all shadow of doubt the first question before the Metallurgical Laboratory; a self-sustaining nuclear chain reaction had been produced in a system using normal uranium. This experiment had been performed under the general direction of E. Fermi, assisted principally by the groups headed by W. H. Zinn and H. L. Anderson. V. C. Wilson and his group had been largely responsible for developing the instruments and controls, and a great many others in the laboratory had contributed to the success of the enterprise.

Relation between Power and Production of Plutonium

6.32. The immediate object of building a uranium-graphite pile was to prove that there were conditions under which a chain reaction would occur, but the ultimate objective of the laboratory was to produce plutonium by a chain reaction. Therefore we are interested in the relation between the power at which a pile operates and the rate at which it produces plutonium. The relation may be evaluated to a first approximation rather easily. A pile running stably must be producing as many neutrons as it is losing. For every thermal neutron absorbed in U-235 a certain number of neutrons, η, is emitted. One of these neutrons is required to maintain the chain. Therefore, assuming the extra neutrons all are absorbed by U-238 to form plutonium, there will be $\eta - 1$ atoms of Pu^{239} formed for every fission. Every fission releases roughly 200 Mev of energy. Therefore the formation of $\eta - 1$ atoms of plutonium accompanies the release of about 200 Mev. Since $\eta - 1$ is a small number, we can guess that to

produce a kilogram a day of plutonium a chain-reacting pile must be releasing energy at the rate of 500,000 to 1,500,000 kilowatts. The first chain-reacting pile that we have just described operated at a maximum of 200 watts. Assuming that a single bomb will require the order of one to 100 kilograms of plutonium, the pile that has been described would have to be kept going at least 70,000 years to produce a single bomb. Evidently the problem of quantity production of plutonium was not yet solved.

The Chemistry of Plutonium

6.33. The second specific objective of the Metallurgical Laboratory was to show that, if a chain reaction did occur, it would be feasible to separate the plutonium chemically from the other material with which it is found. Progress toward this objective was necessarily slower than toward the attainment of a chain reaction. Initially little was done at the Metallurgical Laboratory on chemical problems although the extraction problem was discussed in a conference soon after the project was organized and the work of Seaborg's group at the University of California on plutonium was encouraged. On April 22–23, 1942, a general conference on chemistry was held at Chicago, attended by F. H. Spedding, E. W. Thiele, G. T. Seaborg, J. W. Kennedy, H. C. Urey, E. Wigner, N. Hilberry, G. E. Boyd, I. B. Johns, H. A. Wilhelm, I. Perlman, A. C. Wahl, and J. A. Wheeler. Spedding, in opening the meeting, pointed out that there were two main tasks for the chemists: first, to separate plutonium in the amounts and purity required for war purposes; second, to obtain a good understanding of the chemistry necessary for the construction and maintenance of the pile. The separation problem was to be studied by a new group at Chicago under the direction of Seaborg, by Johns and Wilhelm at Ames, and by Wahl and Kennedy continuing the work at California. Other closely related groups at Chicago were to be C. D. Coryell's, working on the fission products, and Boyd's on analytical problems. The chemistry group at Chicago has grown speedily since that time. A new building had to be constructed to house it late in 1942, and this building was

enlarged subsequently. Altogether, the solving of many of the chemical problems has been one of the most remarkable achievements of the Metallurgical Laboratory.

6.34. The first isotope of plutonium discovered and studied was not the 239 isotope but the 238 isotope, which is an alpha-ray emitter with a half-life of about 50 years. U-238 bombarded with deuterons gives $_{93}Np^{238}$ which disintegrates to $_{94}Pu^{238}$ by beta emission.* The first evidence of the actual existence of these new elements (ruling out the original erroneous interpretation of the splitting of uranium as evidence for their existence) was obtained by E. McMillan and P. H. Abelson who isolated 93-238 from uranium bombarded with deuterons in the Berkeley cyclotron. This new element was identified as a beta emitter but the sample was too small for isolation of the daughter product 94-238. Later, enough Pu-238 was prepared to permit Seaborg, Kennedy and Wahl to begin the study of its chemical properties in the winter of 1940–1941 by using tracer chemistry with carriers according to practice usual in radiochemistry. By such studies many chemical properties of plutonium were determined, and several possible chemical processes were evolved by which Pu-239 might be removed from the chain-reacting pile. The success of experiments on a tracer scale led to plans to produce enough Pu-239 to be treated as an ordinary substance on the ultra-microchemical scale. Such quantities were produced by prolonged bombardment of several hundred pounds of uranyl nitrate with neutrons obtained with the aid of cyclotrons, first at Berkeley and later at Washington University in St. Louis. By the end of 1942, something over 500 micrograms had been obtained in the form of pure plutonium salts. Although this amount is less than would be needed to make the head of a pin, for the micro-chemists it was sufficient to yield considerable information; for one microgram is considered sufficient to carry out weighing experiments, titrations, solubility studies, etc.

6.35. From its position in the periodic table, plutonium might be expected to be similar to the rare earths or to uranium.

* See drawing on p. 8.

thorium, or osmium. Which of these it will resemble most closely depends, of course, on the arrangement of the outermost groups of electrons and this arrangement could hardly have been predicted. On the whole, plutonium turned out to be more like uranium than like any of the other elements named and might even be regarded as the second member of a new rare-earth series beginning with uranium. It was discovered fairly early that there were at least two states of oxidation of plutonium. (It is now known that there are four, corresponding to positive valences of 3, 4, 5, and 6.) Successful microchemical preparation of some plutonium salts and a study of their properties led to the general conclusion that it was possible to separate plutonium chemically from the other materials in the pile. This conclusion represents the attainment of the second immediate objective of the Metallurgical Laboratory. Thus, by the end of 1942, plutonium, entirely unknown eighteen months earlier, was considered an element whose chemical behavior was as well understood as that of several of the elements of the old periodic table.

MISCELLANEOUS STUDIES

6.36. Besides the major problems we have mentioned, i.e., the chain reaction, the chemical separation, and the planning for a production plant, there were innumerable minor problems to be solved. Among the more important of these were the improvement of neutron counters, ionization chambers, and other instruments, the study of corrosion of uranium and aluminum by water and other possible coolants, the determination of the effects of temperature variation on neutron cross sections, the fabrication of uranium rods and tubes, the study of fission products, and the determination of the biological effects of radiation. As typical of this kind of work we can cite the development of methods of fabricating and coating uranium metal, under the direction of E. Creutz. Without the accomplishment of these secondary investigations the project could not have reached its goal. To give some further idea of the scope of the work, a list

of twenty report titles is presented in Appendix 5, the 20 reports being selected from the 400 or so issued during 1942.

THE FAST-NEUTRON REACTION

6.37. The third initial objective of the Metallurgical Project was to obtain theoretical and experimental data on a "fast neutron" reaction, such as would be required in an atomic bomb. This aspect of the work was initially planned and coordinated by G. Breit of the University of Wisconsin and later continued by J. R. Oppenheimer of the University of California. Since the actual construction of the bomb was to be the final part of the program, the urgency of studying such reactions was not so great. Consequently, little attention was given to the theoretical problems until the summer of 1942, when a group was organized at Chicago under the leadership of Oppenheimer.

6.38. In the meantime experimental work initiated in most instances by G. Breit, had been in progress (under the general direction of the Metallurgical Project) at various institutions having equipment suitable for fast-neutron studies (Carnegie Institution of Washington, the National Bureau of Standards, Cornell University, Purdue University, University of Chicago, University of Minnesota, University of Wisconsin, University of California, Stanford University, University of Indiana, and Rice Institute). The problems under investigation involved scattering, absorption and fission cross section, the energy spectrum of fission neutrons, and the time delay in the emission of fission neutrons. For the most part this work represented an intermediate step in confirming and extending previous measurements but reached no new final conclusion. This type of work was subsequently concentrated at another site (see Chapter XII).

6.39. As indicated by the "Feasibility Report" (in a section written by J. H. Manley, J. R. Oppenheimer, R. Serber, and E. Teller) the picture had changed significantly in only one respect since the appearance of the National Academy Report a year earlier. Theoretical studies now showed that the effectiveness of the atomic bomb in producing damage would be greater

than had been indicated in the National Academy report. However, critical size of the bomb was still unknown. Methods of detonating the bomb had been investigated somewhat, but on the whole no certain answers had been reached.

PART II: PROGRESS TOWARD THE ULTIMATE OBJECTIVE

PLANNING A PRODUCTION PLANT

Planning and Technical Work

6.40. As we have seen, the initial objectives of the Metallurgical Laboratory had been reached by the end of 1942, but the ultimate objectives, the production of large quantities of plutonium and the design and fabrication of bombs, were still far from attained. The responsibility for the design and fabrication of bombs was transferred to another group at about this time; its work is reported in Chapter XII. The production of Pu-239 in quantity has remained the principal responsibility of the Metallurgical Laboratory although shared with the du Pont Company since the end of 1942.

6.41. On the basis of the evidence available it was clear that a plutonium production rate somewhere between a kilogram a month and a kilogram a day would be required. At the rate of a kilogram a day, a 500,000 to 1,500,000 kilowatt plant would be required. (The ultimate capacity of the hydroelectric power plants at the Grand Coulee Dam is expected to be 2,000,000 kw.) Evidently the creation of a plutonium production plant of the required size was to be a major enterprise even without attempting to utilize the thermal energy liberated. Nevertheless, by November 1942 most of the problems had been well defined and tentative solutions had been proposed. Although these problems will be discussed in some detail in the next chapter, we will mention them here.

6.42. Since a large amount of heat is generated in any pile producing appreciable amounts of plutonium, the first problem

of design is a cooling system. Before such a system can be designed, it is necessary to find the maximum temperature at which a pile can run safely and the factors—nuclear or structural—which determine this temperature. Another major problem is the method for loading and unloading the uranium, a problem complicated by the shielding and the cooling system. Shielding against radiation has to be planned for both the pile itself and the chemical separation plant. The nature of the separation plant depends on the particular separation process to be used, which has to be decided. Finally, speed of procurement and construction must be primary factors in the planning of both the pile and the chemical plant.

Possible Types of Plant

6.43. After examining the principal factors affecting plant design, i.e., cooling, efficiency, safety, and speed of construction, the "Feasibility Report" suggested a number of possible plant types in the following order of preference:

I. (a) Ordinary uranium metal lattice in a graphite moderator with helium cooling. (b) The same, with water cooling. (c) The same, with molten bismuth cooling.
II. Ordinary uranium metal lattice in a heavy-water moderator.
III. Uranium enriched in the 235 isotope using graphite, heavy water, or ordinary water as moderator.

Types II and III were of no immediate interest since neither enriched uranium nor heavy water was available. Development of both these types continued however, since if no other type proved feasible they might have to be used. Type I (c), calling for liquid bismuth cooling, seemed very promising from the point of view of utilization of the thermal energy released, but it was felt that the technical problems involved could not be solved for a long time.

The Pilot Plant at Clinton

6.44. During this period, the latter half of 1942, when production plants were being planned, it was recognized that a plant of intermediate size was desirable. Such a plant was needed for two reasons: first, as a pilot plant; second, as a producer of a few grams of plutonium badly needed for experimental purposes. Designed as an air-cooled plant of 1,000-kw capacity, the intermediate pile constructed at Clinton, Tennessee, might have served both purposes if helium cooling had been retained for the main plant. Although the plans for the main plant were shifted so that water cooling was called for, the pilot plant was continued with air cooling in the belief that the second objective would be reached more quickly. It thus ceased to be a pilot plant except for chemical separation. Actually the main plant was built without benefit of a true pilot plant, much as if the hydroelectric generators at Grand Coulee had been designed merely from experience gained with a generator of quite different type and of a small fraction of the power.

SPECIFIC PROPOSALS

6.45. As reviewed by Hilberry in the "Feasibility Report" of November 26, 1942, the prospects for a graphite pile with helium cooling looked promising as regards immediate production; the pile using heavy water for moderator and using heavy water or ordinary water as coolant looked better for eventual full-scale use. A number of specific proposals were made for construction of such plants and for the further study of the problems involved. These proposals were based on time and cost estimates which were necessarily little better than rough guesses. As the result of further investigation the actual program of construction—described in later chapters—has been quite different from that proposed.

SUMMARY

6.46. The procurement problem which had been delaying progress was essentially solved by the end of 1942. A small self-

sustaining graphite-uranium pile was constructed in November 1942, and was put into operation for the first time on December 2, 1942, at a power level of ½ watt and later at 200 watts. It was easily controllable thanks to the phenomenon of delayed neutron emission. A total of 500 micrograms of plutonium was made with the cyclotron and separated chemically from the uranium and fission products. Enough was learned of the chemistry of plutonium to indicate the possibility of separation on a relatively large scale. No great advance was made on bomb theory, but calculations were checked and experiments with fast neutrons extended. If anything, the bomb prospects looked more favorable than a year earlier.

6.47. Enough experimenting and planning were done to delineate the problems to be encountered in constructing and operating a large-scale production plant. Some progress was made in choice of type of plant, first choice at that time being a pile of metallic uranium and graphite, cooled either by helium or water. A specific program was drawn up for the construction of pilot and production plants. This program presented time and cost estimates.

CHAPTER VIII. THE PLUTONIUM PROBLEM JANUARY 1943 TO JUNE 1945

INTRODUCTION

8.1. The necessity for pushing the design and construction of the full-scale plutonium plant simultaneously with research and development inevitably led to a certain amount of confusion and inefficiency. It became essential to investigate many alternative processes. It became necessary to investigate all possible causes of failure even when the probability of their becoming serious was very small. Now that the Hanford plant is producing plutonium successfully, we believe it is fair to say that a large percentage of the results of investigation made between the end of 1942 and the end of 1944 will never be used—at least not for the originally intended purposes. Nevertheless had the Hanford plant run into difficulties, any one of the now superfluous investigations might have furnished just the information required to convert failure into success. Even now it is impossible to say that future improvements may not depend on the results of researches that seem unimportant today.

8.2. It is estimated that thirty volumes will be required for a complete report of the significant scientific results of researches conducted under the auspices of the Metallurgical Project. Work was done on every item mentioned on the research program presented in the last chapter. In the present account it would be obviously impossible to give more than a brief abstract of all these researches. We believe this would be unsatisfactory and that it is preferable to give a general discussion of the chain-reacting units and separation plants as they now operate, with some discussion of the earlier developments.

THE CHAIN REACTION IN A PILE

8.3. In Chapter I and other early chapters we have given brief accounts of the fission process, pile operation, and chemical

separation. We shall now review these topics from a somewhat different point of view before describing the plutonium production plants themselves.

8.4. The operation of a pile depends on the passage of neutrons through matter and on the nature of the collisions of neutrons with the nuclei encountered. The collisions of principal importance are the following:

I. Collisions in which neutrons are scattered and lose appreciable amounts of energy. (a) Inelastic collisions of fast neutrons with uranium nuclei. (b) Elastic collisions of fast or moderately fast neutrons with the light nuclei of the moderator material; these collisions serve to reduce the neutron energy to very low (so-called thermal) energies.

II. Collisions in which the neutrons are absorbed. (a) Collisions which result in fission of nuclei and give fission products and additional neutrons. (b) Collisions which result in the formation of new nuclei which subsequently disintegrate radioactively (e.g., ${}_{92}U^{239}$ which produces ${}_{94}Pu^{239}$).

8.5. Only the second class of collision requires further discussion. As regards collisions of Type II (a), the most important in a pile are the collisions between neutrons and U-235, but the high-energy fission of U-238 and the thermal fission of Pu-239 also take place. Collisions of Type II (b) are chiefly those between neutrons and U-238. Such collisions occur for neutrons of all energies, but they are most likely to occur for neutrons whose energies lie in the "resonance" region located somewhat above thermal energies. The sequence of results of the Type II (b) collision is represented as follows:

$$
\begin{aligned}
{}_{92}U^{238} + {}_{0}n^{1} &\longrightarrow {}_{92}U^{239} + \text{gamma rays} \\
{}_{92}U^{239} &\xrightarrow[23\ \text{min.}]{} {}_{93}Np^{239} + {}_{-1}e^{0} \\
{}_{93}Np^{239} &\xrightarrow[2.3\ \text{days}]{} {}_{94}Pu^{239} + {}_{-1}e^{0} + \text{gamma rays}
\end{aligned}
$$

8.6. Any other non-fission absorption processes are important chiefly because they waste neutrons; they occur in the moderator,

in U-235, in the coolant, in the impurities originally present, in the fission products, and even in plutonium itself.

8.7. Since the object of the chain reaction is to generate plutonium, we would like to absorb all excess neutrons in U-238, leaving just enough neutrons to produce fission and thus to maintain the chain reaction. Actually the tendency of the neutrons to be absorbed by the dominant isotope U-238 is so great compared to their tendency to produce fission in the 140-times-rarer U-235 that the principal design effort had to be directed toward favoring the fission (as by using a moderator, a suitable lattice, materials of high purity, etc.,) in order to maintain the chain reaction.

Life History of One Generation of Neutrons*

8.8. All the chain-reacting piles designed by the Metallurgical Laboratory or with its cooperation consist of four categories of material—the uranium metal, the moderator, the coolant, and the auxiliary materials such as water tubes, casings of uranium, control strips or rods, impurities, etc. All the piles depend on stray neutrons from spontaneous fission or cosmic rays to initiate the reaction.

8.9. Suppose that the pile were to be started by simultaneous release (in the uranium metal) of N high-energy neutrons. Most of these neutrons originally have energies above the threshold energy of fission of U-238. However, as the neutrons pass back and forth in the metal and moderator, they suffer numerous inelastic collisions with the uranium and numerous elastic collisions with the moderator, and all these collisions serve to reduce the energies below that threshold. Specifically, in a typical graphite-moderated pile a neutron that has escaped from the uranium into the graphite travels on the average about 2.5 cm between collisions and makes on the *average* about 200 elastic collisions before passing from the graphite back into the uranium. Since at each such collision a neutron loses on the average about one sixth of its energy, a one-Mev neutron is reduced to thermal

* See drawing facing p. 35.

energy (usually taken to be 0.025 electron volt) considerably before completing a single transit through the graphite. There are, of course, many neutrons that depart from this average behavior, and there will be enough fissions produced by fast neutrons to enhance slightly the number of neutrons present. The enhancement may be taken into account by multiplying the original number of neutrons N by a factor ϵ which is called the fast-fission effect or the fast-multiplication factor.

8.10. As the average energy of the $N\epsilon$ neutrons present continues to fall, inelastic collision in the uranium becomes unimportant, the energy being reduced essentially only in the moderator. However, the chance of non-fission absorption (resonance capture) in U-238 becomes significant as the intermediate or resonance energy region is reached. Actually quite a number of neutrons in this energy region will be absorbed regardless of choice of lattice design. The effect of such capture may be expressed by multiplying $N\epsilon$ by a factor p, (which is always less than one) called the "resonance escape probability" which is the probability that a given neutron starting with energy above the resonance region will reach thermal energies without absorption in U-238. Thus from the original N high-energy neutrons we obtain $N\epsilon p$ neutrons of thermal energy.

8.11. Once a neutron has reached thermal energy the chance that it will lose more energy by collision is no greater than the chance that it will gain energy. Consequently the neutrons will remain at this average energy until they are absorbed. In the thermal-energy region the chance for absorption of the neutron by the moderator, the coolant and the auxiliary materials is greater than at higher energies. At any rate it is found that we introduce little error into our calculations by assuming all such unwanted absorption takes place in this energy region. We now introduce a factor f, called the thermal utilization factor, which is defined as the probability that a given thermal neutron will be absorbed in the uranium. Thus from the original N fast neutrons we have obtained $N\epsilon pf$ thermal neutrons which are absorbed by uranium.

8.12. Although there are several ways in which the normal mixture of uranium isotopes can absorb neutrons, the reader may recall that we defined in a previous chapter a quantity η, which is the number of fission neutrons produced for each thermal neutron absorbed in uranium regardless of the details of the process. If, therefore, we multiply the number of thermal neutrons absorbed in uranium, $N\epsilon pf$, by η, we have the number of new high speed neutrons generated by the original N high speed neutrons in the course of their lives. If $N\epsilon pf\eta$ is greater than N, we have a chain reaction and the number of neutrons is continually increasing. Evidently the product $\epsilon pf\eta = k_\infty$, the multiplication factor already defined in Chapter IV.

8.13. Note that no mention has been made of neutrons escaping from the pile. Such mention has been deliberately avoided since the value of k_∞ as defined above applies to an infinite lattice. From the known values of k_∞ and the fact that these piles do operate, one finds that the percentage of neutrons escaping cannot be very great. As we saw in Chapter II, the escape of neutrons becomes relatively less important as the size of the pile increases. If it is necessary to introduce in the pile a large amount of auxiliary material such as cooling-system pipes, it is necessary to build a somewhat larger pile to counteract the increase in absorption.

8.14. To sum up, a pile operates by reducing high-energy neutrons to thermal energies by the use of a moderator-lattice arrangement, then allowing the thermal-energy neutrons to be absorbed by uranium, causing fission which regenerates further high-energy neutrons. The regeneration of neutrons is aided slightly by the fast neutron effect; it is impeded by resonance absorption during the process of energy reduction, by absorption in graphite and other materials, and by neutron escape.

The Effects of Reaction Products on the Multiplication Factor

8.15. Even at the high power level used in the Hanford piles, only a few grams of U-238 and of U-235 are used up per day per

million grams of uranium present. Nevertheless the effects of these changes are very important. As the U-235 is becoming depleted, the concentration of plutonium is increasing. Fortunately, plutonium itself is fissionable by thermal neutrons and so tends to counterbalance the decrease of U-235 as far as maintaining the chain reaction is concerned. However, other fission products are being produced also. These consist typically of unstable and relatively unfamiliar nuclei so that it was originally impossible to predict how great an undesirable effect they would have on the multiplication constant. Such deleterious effects are called poisoning.

THE REACTION PRODUCTS AND THE SEPARATION PROBLEM

8.16. There are two main parts of the plutonium production process at Hanford: actual production in the pile, and separation of the plutonium from the uranium slugs in which it is formed. We turn now to a discussion of the second part, the separation process.

8.17. The uranium slugs containing plutonium also contain other elements resulting from the fission of U-235. When a U-235 nucleus undergoes fission, it emits one or more neutrons and splits into two fragments of comparable size and of total mass 235 or less. Apparently fission into precisely equal masses rarely occurs, the most abundant fragments being a fragment of mass number between 134 and 144 and a fragment of mass number between 100 and 90. Thus there are two groups of fission products: a heavy group with mass numbers extending approximately from 127 to 154, and a light group from approximately 115 to 83. These fission products are in the main unstable isotopes of the thirty or so known elements in these general ranges of mass number. Typically they decay by successive beta emissions accompanied by gamma radiation finally to form known stable nuclei. The half-lives of the various intermediate nuclei range from fractions of a second to a year or more; several of the important species have half-lives of the order of a month or so.

About twenty different elements are present in significant concentration. The most abundant of these comprises slightly less than 10 per cent of the aggregate.

8.18. In addition to radioactive fission products, U-239 and Np-239 (intermediate products in the formation of plutonium) are present in the pile and are radioactive. The concentrations of all these products begin to build up at the moment the pile starts operating. Eventually the rate of radioactive decay equals the rate of formation so that the concentrations become constant. For example, the number of atoms of U-239 produced per second is constant for a pile operating at a fixed power level. According to the laws of radioactive disintegration, the number of U-239 atoms disappearing per second is proportional to the number of such atoms present and is thus increasing during the first few minutes or hours after the pile is put into operation. Consequently there soon will be practically as many nuclei disintegrating each second as are formed each second. Equilibrium concentrations for other nuclei will be approached in similar manner, the equilibrium concentration being proportional to the rate of formation of the nucleus and to its half-life. Products which are stable or of extremely long half-life (e.g., plutonium) will steadily increase in concentration for a considerable time. When the pile is stopped, the radioactivity of course continues, but at a continually diminishing absolute rate. Isotopes of very short half-life may "drop out of sight" in a few minutes or hours; others of longer half-life keep appreciably active for days or months. Thus at any time the concentrations of the various products in a recently stopped pile depend on what the power level was, on how long the pile ran, and on how long it has been shut down. Of course, the longer the pile has run, the larger is the concentration of plutonium and (unfortunately) the larger is the concentration of long-lived fission products. The longer the "cooling" period, i.e., the period between removal of material from the pile and chemical treatment, the lower is the radiation intensity from the fission products. A compromise must be made between such considerations as the desire for a long running and cooling

time on the one hand and the desire for early extraction of the plutonium on the other hand.

8.19. Tables can be prepared showing the chemical concentrations of plutonium and the various fission products as functions of power level, length of operation, and length of cooling period. The half-life of the U-239 is so short that its concentration becomes negligible soon after the pile shuts down. The neptunium becomes converted fairly rapidly to plutonium. Of course, the total weight of fission products, stable and unstable, remains practically constant after the pile is stopped. For the Clinton and Hanford operating conditions the maximum plutonium concentration attained is so small as to add materially to the difficulty of chemical separation.

THE CHOICE OF A CHEMICAL SEPARATION PROCESS

8.20. The problem then is to make a chemical separation at the daily rate of, say, several grams of plutonium from several thousand grams of uranium contaminated with large amounts of dangerously radioactive fission products comprising twenty different elements. The problem is especially difficult as the plutonium purity requirements are very high indeed.

8.21. Four types of method for chemical separation were examined: volatility, absorption, solvent extraction, and precipitation. The work on absorption and solvent extraction methods has been extensive and such methods may be increasingly used in the main process or in waste recovery, but the Hanford Plant was designed for a precipitation process.

8.22.* The phenomena of co-precipitation, i.e., the precipitation of small concentrations of one element along with a "carrier" precipitate of some other element, had been commonly used in radioactive chemistry, and was adopted for plutonium separation. The early work on plutonium chemistry, confined as it was to minute amounts of the element, made great use of precipitation

* Paragraphs 8.22–8.26 are quoted or paraphrased from a general report of the Metallurgical Laboratory prepared in the spring of 1945.

reactions from which solubility properties could be deduced. It was therefore natural that precipitation methods of separation were the most advanced at the time when the plant design was started. It was felt that, should the several steps in the separations process have to be developed partly by the empirical approach, there would be less risk in the scale-up of a precipitation process than, for example, of one involving solid-phase reactions. In addition, the precipitation processes then in mind could be broken into a sequence of repeated operations (called cycles), thereby limiting the number of different equipment pieces requiring design and allowing considerable process change without equipment change. Thus, while the basic plant design was made with one method in mind, the final choice of a different method led to no embarrassments.

8.23. Most of the precipitation processes which have received serious consideration made use of an alternation between the (IV) and (VI) oxidation states of plutonium. Such processes involve a precipitation of plutonium (IV) with a certain compound as a carrier, then dissolution of the precipitate, oxidation of the plutonium to the (VI) state, and reprecipitation of the carrier compound while the plutonium (VI) remains in solution. Fission products which are not carried by these compounds remain in solution when plutonium (IV) is precipitated. The fission products which carry are removed from the plutonium when it is in the (VI) state. Successive oxidation-reduction cycles are carried out until the desired decontamination is achieved. The process of elimination of the fission products is called decontamination and the degree of elimination is tested by measuring the change in radioactivity of the material.

Combination Processes

8.24. It is possible to combine or couple the various types of process. Some advantages may be gained in this way since one type of process may supplement another. For example, a process which gives good decontamination might be combined advan-

tageously with one which, while inefficient for decontamination, would be very efficient for separation from uranium.

8.25. At the time when it became necessary to decide on the process to serve as the basis for the design of the Hanford plant (June 1943), the choice, for reasons given above, was limited to precipitation processes and clearly lay between two such processes. However, the process as finally chosen actually represented a combination of the two.

8.26. The success of the separation process at Hanford has exceeded all expectations. The high yields and decontamination factors and the relative ease of operation have amply demonstrated the wisdom of its choice as a process. This choice was based on a knowledge of plutonium chemistry which had been gleaned from less than a milligram of plutonium. Further developments may make the present Hanford process obsolete, but the principal goal, which was to have a workable and efficient process for use as soon as the Hanford piles were delivering plutonium, has been attained.

THE ARGONNE LABORATORY

8.27. The Argonne Laboratory was constructed early in 1943 outside Chicago. The site, originally intended for a pilot plant, was later considered to be too near the city and was used for reconstructing the so-called West Stands pile which was originally built on the University of Chicago grounds and which was certainly innocuous. Under the direction of E. Fermi and his colleagues, H. L. Anderson, W. H. Zinn, G. Weil, and others, this pile has served as a prototype unit for studies of thermal stability, controls, instruments, and shielding, and as a neutron source for materials testing and neutron-physics studies. Furthermore, it has proved valuable as a training school for plant operators. More recently a heavy-water pile (see below) has been constructed there.

8.28. The first Argonne pile, a graphite-uranium pile, need not be described in detail. The materials and lattice structure are nearly identical to those which were used for the original

West Stands pile. The pile is a cube; it is surrounded by a shield and has controls and safety devices somewhat similar to those used later at Clinton. It has no cooling system and is normally run at a power level of only a few kilowatts. It has occasionally been run at high-power levels for very brief periods. Considering that it is merely a reconstruction of the first chain-reacting unit ever built, it is amazing that it has continued in operation for more than two years without developing any major troubles.

8.29. One of the most valuable uses of the Argonne pile has been the measurement of neutron-absorption cross sections of a great variety of elements which might be used in piles as structural members, etc., or which might be present in pile materials as impurities. These measurements are made by observing the change in the controls necessary to make k_{eff} equal to 1.00 when a known amount of the substance under study is inserted at a definite position in the pile. The results obtained were usually expressed in terms of "danger coefficients."

8.30. An opening at the top of the pile lets out a very uniform beam of thermal neutrons that can be used for exponential-pile experiments, for direct measurements of absorption cross sections, for Wilson cloud chamber studies, etc.

8.31. An interesting phenomenon occurring at the top of the pile is the production of a beam or flow of "cold" neutrons. If a sufficient amount of graphite is interposed between the upper surface of the pile and an observation point a few yards above, the neutron energy distribution is found to correspond to a temperature much lower than that of the graphite. This is presumed to be the result of a preferential transmission by the crystalline graphite of the slowest ("coldest") neutrons, whose quantum-mechanical wave-length is great compared to the distance between successive planes in the graphite crystals.

8.32. More recently a pile using heavy water as moderator was constructed in the Argonne Laboratory. The very high intensity beam of neutrons produced by this pile has been found well-suited to the study of "neutron optics," e.g., reflection and refraction of neutron beams as by graphite.

8.33. A constant objective of the Argonne Laboratory has been a better understanding of nuclear processes in uranium, neptunium, and plutonium. Repeated experiments have been made to improve the accuracy of constants such as thermal-fission cross sections of U-235, U-238, and Pu-239, probabilities of non-fission neutron absorption by each of these nuclei, and number of neutrons emitted per fission.

THE CLINTON PLANT

8.34. In Chapter VI we mentioned plans for a "pilot" plant for production of plutonium to be built at the Clinton site in Tennessee. By January 1943, the plans for this project were well along; construction was started soon afterward. M. D. Whitaker was appointed director of the Clinton Laboratories. The pilot-plant plans were made cooperatively by du Pont and the Metallurgical Laboratory; construction was carried out by du Pont; plant operation was maintained by the University of Chicago as part of the Metallurgical Project.

8.35. The main purposes of the Clinton plant were to produce some plutonium and to serve as a pilot plant for chemical separation. As regards research, the emphasis at Clinton was on chemistry and on the biological effects of radiations. A large laboratory was provided for chemical analysis, for research on purification methods, for fission-product studies, for development of intermediate-scale extraction and decontamination processes, etc. Later a "hot laboratory," i.e., a laboratory for remotely-controlled work on highly radioactive material, was provided. There is also an instrument shop and laboratory that has been used very actively. There are facilities for both clinical and experimental work of the health division, which has been very active. There is a small physics laboratory in which some important work was done using higher neutron intensities than were available at the Argonne Laboratory. The principal installations constructed at the Clinton Laboratory site were the pile and the separation plant; these are briefly described below.

The Clinton Pile

8.36. In any steadily operating pile the effective multiplication factor k must be kept at 1, whatever the power level. The best k_∞ that had been observed in a uranium-graphite lattice could not be achieved in a practical pile because of neutron leakage, cooling system, cylindrical channels for the uranium, protective coating on the uranium, and other minor factors. Granted air-cooling and a maximum safe temperature for the surface of the uranium, a size of pile had to be chosen that could produce 1,000 kw. The effective k would go down with rising temperature but not sufficiently to be a determining factor. Though a sphere was the ideal shape, practical considerations recommended a rectangular block.

8.37. The Clinton pile consists of a cube of graphite containing horizontal channels filled with uranium. The uranium is in the form of metal cylinders protected by gas-tight casings of aluminum. The uranium cylinders or slugs may be slid into the channels in the graphite; space is left to permit cooling air to flow past, and to permit pushing the slugs out at the back of the pile when they are ready for processing. Besides the channels for slugs there are various other holes through the pile for control rods, instruments, etc.

8.38. The Clinton pile was considerably larger than the first pile at Chicago (see Chapter VI). More important than the increased size of the Clinton pile were its cooling system, heavier shields, and means for changing the slugs. The production goal of the Clinton plant was set at a figure which meant that the pile should operate at a power level of 1,000 kw.

8.39. The instrumentation and controls are identical in principle to those of the first pile. Neutron intensity in the pile is measured by a BF_3 ionization chamber and is controlled by boron steel rods that can be moved in and out of the pile, thereby varying the fraction of neutrons available to produce fission.

8.40. In spite of an impressive array of instruments and safety devices, the most striking feature of the pile is the simplicity of

operation. Most of the time the operators have nothing to do except record the readings of various instruments.

The Separation Plant

8.41. Here, as at Hanford, the plutonium processes have to be carried out by remote control and behind thick shields. The separation equipment is housed in a series of adjacent cells having heavy concrete walls. These cells form a continuous structure (canyon) which is about 100 feet long and is two-thirds buried in the ground. Adjacent to this canyon are the control rooms, analytical laboratories, and a laboratory for further purification of the plutonium after it has been decontaminated to the point of comparative safety.

8.42. Uranium slugs that have been exposed in the pile are transferred under water to the first of these cells and are then dissolved. Subsequent operations are performed by pumping solutions or slurries from one tank or centrifuge to another.

Performance of Clinton Pile

8.43. The Clinton pile started operating on November 4, 1943, and within a few days was brought up to a power level of 500 kw at a maximum slug surface temperature of 110° C. Improvements in the air circulation and an elevation of the maximum uranium surface temperature to 150° C. brought the power level up to about 800 kw, where it was maintained until the spring of 1944. Starting at that time, a change was made in the distribution of uranium, the change being designed to level out the power distribution in the pile by reducing the amount of metal near the center relative to that further out and thereby to increase the average power level without anywhere attaining too high a temperature. At the same time improvements were realized in the sealing of the slug jackets, making it possible to operate the pile at higher temperature. As a result, a power level of 1,800 kw was attained in May 1944; this was further increased after the installation of better fans in June 1944.

8.44. Thus the pile performance of June 1944 considerably exceeded expectations. In ease of control, steadiness of operation, and absence of dangerous radiation, the pile has been most satisfactory. There have been very few failures attributable to mistakes in design or construction.

8.45. The pile itself was simple both in principle and in practice. Not so the plutonium-separation plant. The step from the first chain-reacting pile to the Clinton pile was reasonably predictable; but a much greater and more uncertain step was required in the case of the separation process, for the Clinton separation plant was designed on the basis of experiments using only microgram amounts of plutonium.

8.46. Nevertheless, the separation process worked! The first batch of slugs from the pile entered the separation plant on December 20, 1943. By the end of January 1944, metal from the pile was going to the separation plant at the rate of $\frac{1}{3}$ ton per day. By February 1, 1944, 190 mg of plutonium had been delivered and by March 1, 1944, several grams had been delivered. Furthermore, the efficiency of recovery at the very start was about 50 per cent, and by June 1944 it was between 80 and 90 per cent.

8.47. During this whole period there was a large group of chemists at Clinton working on improving the process and developing it for Hanford. The Hanford problem differed from that at Clinton in that much higher concentrations of plutonium were expected. Furthermore, though the chemists were to be congratulated on the success of the Clinton plant, the process was complicated and expensive. Any improvements in yield or decontamination or in general simplification were very much to be sought.

8.48. Besides the proving of the pile and the separation plant and the production of several grams of plutonium for experimental use at Chicago, Clinton, and elsewhere, the Clinton Laboratories have been invaluable as a training and testing center for Hanford, for medical experiments, pile studies, purification studies, and physical and chemical studies of plutonium and fission products.

8.49. As typical of the kind of problems tackled there and at

Chicago, the following problems—listed in a single routine report for May 1944—are pertinent:

Problems Closed Out during May 1944: Search for New Oxidizing Agent, Effect of Radiation on Water and Aqueous Solutions, Solubility of Plutonium Peroxide, Plutonium Compounds Suitable for Shipment, Fission Product Distribution in Plant Process Solutions, Preliminary Process Design for Adsorption Extraction, Adsorption Semi-Works Assistance, Completion of Adsorption Process Design.

New Problems Assigned during May 1944: New Product Analysis Method, Effect of Radiation on Graphite, Improvement in Yield, New Pile Explorations, Waste Uranium Recovery, Monitoring Stack Gases, Disposal of Active Waste Solutions, Spray Cooling of X Pile, Assay Training Program, Standardization of Assay Methods, Development of Assay Methods, Shielded Apparatus for Process Control Assays, Cloud Chamber Experiment, Alpha Particles from U-235, Radial Product Distribution, Diffraction of Neutrons.

THE HANFORD PLANT

8.50. It is beyond the scope of this report to give any account of the construction of the Hanford Engineer Works, but it is to be hoped that the full story of this extraordinary enterprise and the companion one, the Clinton Engineer Works, will be published at some time in the future. The Hanford site was examined by representatives of General Groves and of du Pont at the end of 1942, and use of the site was approved by General Groves after he had inspected it personally. It was on the west side of the Columbia River in central Washington north of Pasco. In the early months of 1943 a 200-square-mile tract in this region was acquired by the government (by lease or purchase) through the Real Estate Division of the Office of the Chief of Engineers. Eventually an area of nearly a thousand square miles was brought under government control. At the time of acquisition of the land there were a few farms and two small villages, Hanford and Richland, on the site, which was otherwise sage-brush plains

and barren hills. On the 6th of April, 1943, ground was broken for the Hanford construction camp. At the peak of activity in 1944, this camp was a city of 60,000 inhabitants, the fourth largest city in the state. Now, however, the camp is practically deserted as the operating crew is housed at Richland.

8.51. Work was begun on the first of the Hanford production piles on June 7, 1943, and operation of the first pile began in September 1944. The site was originally laid out for five piles, but the construction of only three has been undertaken. Besides the piles, there are, of course, plutonium separation plants, pumping stations and water-treatment plants. There is also a low-power chain-reacting pile for material testing. Not only are the piles themselves widely spaced for safety—several miles apart —but the separation plants are well away from the piles and from each other. All three piles were in operation by the summer of 1945.

Canning and Corrosion

8.52. No one who lived through the period of design and construction of the Hanford plant is likely to forget the "canning" problem, i.e., the problem of sealing the uranium slugs in protective metal jackets. On periodic visits to Chicago the writer could roughly estimate the state of the canning problem by the atmosphere of gloom or joy to be found around the laboratory. It was definitely not a simple matter to find a sheath that would protect uranium from water corrosion, would keep fission products out of the water, would transmit heat from the uranium to the water, and would not absorb too many neutrons. Yet the failure of a single can might conceivably require shut-down of an entire operating pile.

8.53. Attempts to meet the stringent requirements involved experimental work on electroplating processes, hot-dipping processes, cementation-coating processes, corrosion-resistant alloys of uranium, and mechanical jacketing or canning processes. Mechanical jackets or cans of thin aluminum were feasible from the nuclear-physics point of view and were chosen early as the

most likely solution of the problem. But the problem of getting a uniform, heat-conducting bond between the uranium and the surrounding aluminum, and the problem of effecting a gas-tight closure for the can both proved very troublesome. Development of alternative methods had to be carried along up to the last minute, and even up to a few weeks before it was time to load the uranium slugs into the pile there was no certainty that any of the processes under development would be satisfactory. A final minor but apparently important modification in the preferred canning process was adopted in October 1944, after the first pile had begun experimental operation. By the summer of 1945, there had been no can failure reported.

Present Status of the Hanford Plants

8.54. During the fall of 1944 and the early months of 1945 the second and third Hanford piles were finished and put into operation, as were the additional chemical separation plants. There were, of course, some difficulties; however, none of the fears expressed as to canning failure, film formation in the water tubes, or radiation effects in the chemical processes, have turned out to be justified. As of early summer 1945 the piles are operating at designed power, producing plutonium, and heating the Columbia River.* The chemical plants are separating the plutonium from the uranium and from the fission products with better efficiency than had been anticipated. The finished product is being delivered. How it can be used is the subject of Chapter XII.

THE WORK ON HEAVY WATER

8.55. In previous chapters there have been references to the advantages of heavy water as a moderator. It is more effective than graphite in slowing down neutrons and it has a smaller neutron absorption than graphite. It is therefore possible to build

*The actual rise in temperature is so tiny that no effect on fish life could be expected. To make doubly sure, this expectation was confirmed by an elaborate series of experiments.

a chain-reacting unit with uranium and heavy water and thereby to attain a considerably higher multiplication factor, k, and a smaller size than is possible with graphite. But one must have the heavy water.

8.56. In the spring of 1943 the Metallurgical Laboratory decided to increase the emphasis on experiments and calculations aimed at a heavy-water pile. To this end a committee was set up under E. Wigner, a group under H. C. Vernon was transferred from Columbia to Chicago, and H. D. Smyth, who had just become associate director of the Laboratory, was asked to take general charge.

8.57. The first function of this group was to consider in what way heavy water could best be used to insure the overall success of the Metallurgical Project, taking account of the limited production schedule for heavy water that had been already authorized.

8.58. It became apparent that the production schedule was so low that it would take two years to produce enough heavy water to "moderate" a fair-sized pile for plutonium production. On the other hand, there might be enough heavy water to moderate a small "laboratory" pile, which could furnish information that might be valuable. In any event, during the summer of 1943 so great were the uncertainties as to the length of the war and as to the success of the other parts of the DSM project that a complete study of the possibilities of heavy-water piles seemed desirable. Either the heavy-water production schedule might be stepped up or the smaller, experimental pile might be built. An intensive study of the matter was made during the summer of 1943 but in November it was decided to curtail the program and construction was limited to a 250-kw pile located at the Argonne site.

The Argonne Heavy-Water Pile

8.59. Perhaps the most striking aspect of the uranium and heavy-water pile at the Argonne is its small size. Even with its surrounding shield of concrete it is relatively small compared to the uranium-graphite piles.

8.60. By May 15, 1944, the Argonne uranium and heavy-water pile was ready for test. With the uranium slugs in place, it was found that the chain reaction in the pile became self sustaining when only three fifths of the heavy water had been added. The reactivity of the pile was so far above expectations that it would have been beyond the capacity of the control rods to handle if the remainder of the heavy water had been added. To meet this unusual and pleasant situation some of the uranium was removed and extra control rods were added.

8.61. With these modifications it was possible to fill the tank to the level planned. By July 4, 1944, W. H. Zinn reported that the pile was running satisfactorily at 190 kw, and by August 8, 1944, he reported that it was operating at 300 kw.

8.62. In general the characteristics of this pile differed slightly from those of comparable graphite piles. This pile takes several hours to reach equilibrium. It shows small (less than 1 per cent) but sudden fluctuations in power level, probably caused by bubbles in the water. It cannot be shut down as completely or as rapidly as the graphite pile because of the tendency of delayed gamma rays to produce (from the heavy water) additional neutrons. As anticipated, the neutron density at the center is high. The shields, controls, heat exchanger, etc., have operated satisfactorily.

THE HEALTH DIVISION

8.63. The major objective of the health group was in a sense a negative one, to insure that no one concerned suffered serious injury from the peculiar hazards of the enterprise. Medical case histories of persons suffering serious injury or death resulting from radiation were emphatically not wanted. The success of the health division in meeting these problems was remarkable. Even in the research group where control is most difficult, cases showing even temporary bad effects were extremely rare. Factors of safety used in plant design and operation are so great that the hazards of the home and the family car are far greater for the personnel than any arising from the plants.

8.64. To achieve its objective the health group worked along three major lines:

(1) Adoption of pre-employment physical examinations and frequent re-examinations, particularly of those exposed to radiation.

(2) Setting of tolerance standards for radiation doses and development of instruments measuring exposure of personnel; giving advice on shielding, etc.; continually measuring radiation intensities at various locations in the plants; measuring contamination of clothes, laboratory desks, waste water, the atmosphere, etc.

(3) Carrying out research on the effects of direct exposure of persons and animals to various types of radiation, and on the effects of ingestion and inhalation of the various radioactive or toxic materials such as fission products, plutonium and uranium.

Routine Examinations

8.65. The white blood-corpuscle count was used as the principal criterion as to whether a person suffered from overexposure to radiation. A number of cases of abnormally low counts were observed and correlated with the degree of overexposure. Individuals appreciably affected were shifted to other jobs or given brief vacations; none has shown permanent ill effects.

8.66. At the same time it was recognized that the white blood-corpuscle count is not an entirely reliable criterion. Some work on animals indicated that serious damage might occur before the blood count gave any indication of danger. Accordingly, more elaborate blood tests were made on selected individuals and on experimental animals in the hope of finding a test that would give an earlier warning of impending injury.

Instruments for Radiation Measurements

8.67. The Health Division had principal responsibility for the development of pocket meters for indicating the extent of exposure of persons. The first of these instruments was a simple

electroscope about the size and shape of a fountain pen. Such instruments were electrostatically charged at the start of each day and were read at the end of the day. The degree to which they became discharged indicated the total amount of ionizing radiation to which they had been exposed. Unfortunately they were none too rugged and reliable, but the error of reading was nearly always in the right direction—i.e., in the direction of overstating the exposure. At an early date the practice was established of issuing two of these pocket meters to everyone entering a dangerous area. A record was kept of the readings at the time of issuance and also when the meters were turned in. The meters themselves were continually although gradually improved. The Health Division later introduced "film badges," small pieces of film worn in the identification badge, the films being periodically developed and examined for radiation blackening. These instruments for individuals such as the pocket meter and film badge were extra and probably unnecessary precautions. In permanent installations the shielding alone normally affords complete safety. Its effect is under frequent survey by either permanently installed or portable instruments.

8.68. The Health Division cooperated with the Physics Division in the development and use of various other instruments. There was "Sneezy" for measuring the concentration of radioactive dust in the air and "Pluto" for measuring α-emitting contamination (usually plutonium) of laboratory desks and equipment. Counters were used to check the contamination of laboratory coats before and after the coats were laundered. At the exit gates of certain laboratories concealed counters sounded an alarm when someone passed whose clothing, skin or hair was contaminated. In addition, routine inspections of laboratory areas were made.

8.69. One of the studies made involved meteorology. It became essential to know whether the stack gases (at Clinton and at Hanford) would be likely to spread radioactive fission products in dangerous concentrations. Since the behavior of these gases is very dependent on the weather, studies were made at both

sites over a period of many months, and satisfactory stack operation was specified.

Research

8.70. Since both the scale and the variety of the radiation hazards in this enterprise were unprecedented, all reasonable precautions were taken; but no sure means were at hand for determining the adequacy of the precautions. It was essential to supplement previous knowledge as completely as possible. For this purpose, an extensive program of animal experimentation was carried out along three main lines: (1) exposure to neutron, alpha, beta and gamma radiation; (2) ingestion of uranium, plutonium and fission products; (3) inhalation of uranium, plutonium and fission products. Under the general direction of Dr. Stone these experiments were carried out at Chicago, Clinton and the University of California principally by Dr. Cole and Dr. Hamilton. Extensive and valuable results were obtained.

SUMMARY

8.71. Both space and security restrictions prevent a detailed report on the work of the laboratories and plants concerned with plutonium production.

8.72. Two types of neutron absorption are fundamental to the operation of the plant: one, neutron absorption in U-235 resulting in fission, maintains the chain reaction as a source of neutrons; the other, neutron absorption in U-238 leads to the formation of plutonium, the desired product.

8.73. The course of a nuclear chain reaction in a graphite-moderated heterogeneous pile can be described by following a single generation of neutrons. The original fast neutrons are slightly increased in number by fast fission, reduced by resonance absorption in U-238 and further reduced by absorption at thermal energies in graphite and other materials and by escape; the remaining neutrons, which have been slowed in the graphite, cause fission in U-235, producing a new generation of fast neutrons similar to the previous generation.

8.74. The product, plutonium, must be separated by chemical processes from a comparable quantity of fission products and a much larger quantity of uranium. Of several possible separation processes the one chosen consists of a series of reactions including precipitating with carriers, dissolving, oxidizing and reducing.

8.75. The chain reaction was studied at low power at the Argonne Laboratory beginning early in 1943. Both chain reaction and chemical separation processes were investigated at the Clinton Laboratories beginning in November 1943, and an appreciable amount of plutonium was produced there.

8.76. Construction of the main production plant at Hanford, Washington, was begun in 1943 and the first large pile went into operation in September 1944. The entire plant was in operation by the summer of 1945 with all chain-reacting piles and chemical-separation plants performing better than had been anticipated.

8.77. Extensive studies were made on the use of heavy water as a moderator and an experimental pile containing heavy water was built at the Argonne Laboratory. Plans for a production plant using heavy water were given up.

8.78. The Health Division was active along three main lines: (1) medical examination of personnel; (2) advice on radiation hazards and constant check on working conditions; (3) research on the effects of radiation. The careful planning and exhaustive research work of this division have resulted in an outstanding health record at Hanford and elsewhere in the project.