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MANHATTAN DISTRICT HISTORY

BOOK I - GENERAL

VOLUME 4 - AUXILIARY ACTIVITIES

CHAPTER 12 - ACTIVITIES OF

THE NATIONAL BUREAU OF STANDARDS

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THE NATIONAL BUREAU OF STANDARDS

1. General. Undoubtedly the most spectacular work undertaken by the National Bureau of Standards was that for the atomic bomb project. The large scope of the project during its development and engineering phases, as well as the necessary secrecy associated with the program, tends to obscure the initial and critical two years during which the major program was formulated and much basic research was performed. It was in that period that the National Bureau of Standards played a key role in the project, for shortly after the initial phase was completed, the Manhattan District was established and the gigantic development and engineering of the weapon began.

However, atomic project activity by the Bureau did not terminate with the establishment of the Manhattan District. Bureau of Standards experience, staff and facility resources, gained through years of research in atomic and sub-atomic physics, particularly in the field of naturally radioactive substances, served to a great extent in solving many problems throughout all phases of the project. It is, therefore, the purpose of this chapter to recount briefly the outstanding atomic bomb project contributions which were made by the National Bureau of Standards up to the termination of the Manhattan District on 31 December 1946.

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2. Initiation of the Project. The National Bureau of Standards began its work on the atomic bomb project in October 1939 when President Roosevelt appointed its Director, Lyman J. Briggs, as chairman of an Advisory Committee on Uranium to investigate the possibility of utilizing the atomic fission in warfare. At that time it was not known which of the isotopes of uranium was subject to fission when bombarded with slow neutrons, or how many neutrons were emitted in the fission process. The possibility of establishing a chain reaction remained to be demonstrated. Little was known about the chemistry and metallurgy of uranium. Physical methods for separating the isotopes of uranium had yet to be developed.

Action toward the solution of such problems was undertaken by the committee with the enthusiastic cooperation of leading scientists in the respective fields. The uranium committee recognized the future possibilities of uranium fission as a source of power, but, early, decided to concentrate its activities on the development of an atomic bomb. The basic information gained in the initial study could later be applied in the development of atomic power.

Owing to the secrecy imposed on the project, the acquirement of necessary funds became a problem in itself. The experiments begun in 1939 were financed by a transfer of \$6,000 from the Ordnance Department of the Army and the Bureau of Ordnance of the Navy. That money was used to purchase materials for experiments, by Dr. E. Fermi at Columbia University, to determine the suitability of graphite as a moderator to slow down fast neutrons. In 1940, through the cooperation and enthusiastic support of Rear Admiral H. G. Bowen, Director of the Naval Research

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Laboratory, funds to the extent of \$100,000 were made available to the project at a time when they were most urgently needed.

These funds were used mainly for the study of different methods of separating the isotopes. Among them was the method of thermal diffusion proposed by P. H. Abelson, who demonstrated its possibilities in his laboratory at the National Bureau of Standards, carried it successfully through the pilot plant stage at the Naval Research Laboratory, and finally saw it culminate in an operating installation at Oak Ridge, Tennessee. Work was also started at Columbia University by J. R. Dunning on the separation of the uranium isotopes by gaseous diffusion through a porous membrane, which, likewise, eventually led to the construction and operation of an enormous diffusion plant at Oak Ridge. The development of a centrifugal method for separating the isotopes was undertaken by J. W. Beams at the University of Virginia. That method was carried through the pilot plant stage, with highly satisfactory yields, but was not finally selected as a production method.

In June, 1940, President Roosevelt established the National Defense Research Committee under the direction of Dr. Vannevar Bush and made the Uranium Committee a section of NDRC. This was a happy solution to the problem of securing the necessary funds for a secret project, as it was recognized at the outset that the NDRC would be engaged in highly confidential work. At that time the Uranium (S-1) Section was reorganized as follows: L. J. Briggs (chairman), G. B. Pegram (vice-chairman), H. C. Urey, J. W. Beams, M. A. Tuve, R. Gunn, and G. Breit. Additional support was given by the NDRC to the investigations already under way, and new work was undertaken.

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The possibility of establishing a chain reaction was still a matter of great uncertainty, yet upon this the success of the whole project rested. Forty tons of graphite and seven tons of uranium oxide were supplied to Fermi at Columbia for use in a further attack on this problem. New projects dealing with the chain reaction were assigned to the Universities of Chicago, California and Princeton. E. O. Lawrence reported in May, 1941, on experiments at Berkeley which indicated that plutonium (element 94) is formed from uranium 238 (element 92) by neutron capture followed by two beta-transformations and that plutonium like uranium 235 undergoes fission when it captures a slow neutron. This significant discovery held great possibilities because; (1) the abundant isotope 238 could be utilized, (2) separation of the uranium isotopes by physical methods was unnecessary, and (3) plutonium could be separated from the parent mass by chemical methods.

In July, 1941, the membership of the S-1 Section of NDRC was changed somewhat to provide representation for new activities. The meetings, as before, were held at the National Bureau of Standards and were attended by Dr. J. B. Conant as Dr. Bush's representative.

In the spring of 1941 Briggs asked Bush to appoint an independent committee to make an impartial review of the project. Bush requested the National Academy of Sciences to undertake this study and to recommend the level of expenditure at which the investigation should be continued. Up to that time, the expenditures for the atomic bomb project had certainly been moderate. At a meeting of the NDRC on July 18, 1941, Briggs, as chairman of the S-1 Section, reviewed the progress that had been made, and, on the basis of the National Academy of Sciences' reports,

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presented the request of his section for an allotment of \$267,000, with the statement that much larger expenditures would probably soon be required. That allotment was approved by NDRC and theoretical and experimental work went forward at an increased tempo.

In November 1941 Bush, after reviewing the further advances that had been made by the S-1 Section and additional recommendations of the National Academy of Sciences Committee and after consultation with the President and his advisors, decided that the time had now come for an "all-out" effort. The project was transferred to the Office of Scientific Research and Development as an OSRD S-1 Section, which was organized as follows: L. J. Briggs (chairman), G. B. Pegram (vice-chairman), and J. B. Conant; A. H. Compton, E. O. Lawrence, and H. C. Urey, program chiefs; E. V. Murphree, chairman of the planning board; S. K. Allison, J. W. Beams, G. Breit, E. U. Condon, and H. D. Smyth. H. T. Wensel served as technical aide to the committee.

Funds available for the work were greatly expanded. Under Lawrence the investigation of the large-scale separation of the uranium isotopes by electromagnetic processes was accelerated at the University of California. Research on the gaseous diffusion process of separating the isotopes, and on the all important problem of developing suitable porous membranes that would not corrode and become clogged through the action of uranium hexafluoride, was continued at Columbia University under Dunning and Pegram. Urey had supervision of methods for producing heavy water for use as a moderator and other important special assignments. Basic work relating to the chain reaction and its utilization in the production of plutonium, at Columbia under Fermi, and at

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Chicago under Allison, was continued under Compton's direction at the University of Chicago. It was here that the chain-reaction was first demonstrated by Fermi in a graphite-uranium pile in December 1942.

By July, 1942, it appeared that the final intensive work on the research and development program could be directed more effectively by a smaller group and the S-1 Section was succeeded by the S-1 Executive Committee, consisting of Conant, chairman, Briggs, Compton, Lawrence, Murphree and Urey, with Wansel as technical aide and Irvin Stewart as secretary. Committee meetings were held at the Carnegie Institution and were regularly attended by Brigadier General L. R. Groves, head of the recently formed Manhattan District project, who was charged with the heavy responsibility for erecting plants and producing uranium 235 and plutonium in usable quantities by methods developed through the S-1 Committee research groups. By May 1, 1943, the undertaking had reached the point where it could be taken over advantageously by the Manhattan District and the work of OSRD on the uranium project was terminated.

The foregoing is presented in somewhat greater detail in Book I, Volume 1, of the Manhattan District History; details have also been presented in the report "Atomic Energy for Military Purposes" (Smyth Report) (App. 31a of Book I, Volume 4, Chapter 5, M.D. History). The information is repeated in this chapter, however, in order to emphasize the important roll enacted by the Director of the National Bureau of Standards in the initiation of the atomic bomb project.

3. Specific Activities. During the war about sixty members of the Bureau staff were engaged on the atomic bomb project. Some of its leading physicists, chemists and mathematicians were assigned to the Clinton

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and Los Alamos Laboratories at the request of the War Department.

Early in the war a simple and highly effective chemical method was developed at the Bureau for removing harmful impurities from uranium. This was used in all subsequent production of uranium. Similarly a procedure was developed for producing graphite practically free from boron. These contributions were of great value to the project, because some of these impurities were strong absorbers of neutrons and their effective removal was a prerequisite to the establishment of a chain reaction in a pile using normal uranium.

The Bureau also served throughout the war as a central control laboratory for determining the purity of the uranium and other products that were used in the project. Thousands of chemical and spectrographic analyses were made during the course of this work. Measurements were made of the radium recovered from the uranium ores. Methods for separating the uranium isotopes were studied. The alpha-particle counting method was developed to measure the change in the relative abundance of the uranium isotopes during separation processes. Yet the work was so closely guarded that the Bureau's participation in the atomic bomb project was not known to the members of the staff not associated with the undertaking.

a. The Graphite Problem. In the early consideration of the use of atomic energy for military purposes, one of the first proposals (by Fermi and L. Szilard) was to use graphite as a moderator in obtaining a controllable chain reaction for the production of plutonium. It was known that impurities which might be present in the graphite or uranium would capture some of the neutrons generated by fission of the uranium

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235, probably a large enough proportion of them to prevent a self-sustaining reaction from occurring in a uranium-graphite pile. Thus the problems of procuring sufficiently pure graphite and sufficiently pure uranium were outstandingly important among the many assignments undertaken by the Bureau in connection with the atomic energy project.

While graphite was a common article of commerce, its usefulness had never been seriously affected by the impurities that now assumed such commanding importance. In January, 1941, L. Szilard and D. P. Mitchell of Columbia University came to the Bureau to discuss possible reasons for the discrepancy between the analysis of graphite ash, made by a commercial analyst (by a spectrographic method), and the neutron absorption measurements of the ash, made by E. Fermi. It was concluded that the analytical results, for boron, in particular, were at fault. Hence, the establishment of a reliable method for the determination of boron in graphite was undertaken by C. J. Rodden.

The procedure developed, which became standard for the determination of boron in uranium compounds as well as in graphite, showed that much of the neutron absorption of the graphite ash measured by Fermi was due to boron.

With a reliable method for determining small amounts of boron as a guide, the next step was to find out the sources of this impurity. A painstaking survey by the Bureau, of the boron content of the raw materials used in manufacturing graphite and of the distribution of boron in the finished bars, disclosed that the foundry coke universally used in one stage of the graphitizing process was the chief source of the boron contamination. Analyses showed that foundry coke contained about 15

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times as much boron as the petroleum coke then available. It was also found that the surface of the finished graphite bars contained more boron than the inner portion. A thorough canvass of the graphite industry was made and the Speer Carbon Company, St. Marys, Pennsylvania, agreed to substitute petroleum coke for foundry coke and to machine the surfaces of the finished product. The graphite which was then obtained was much lower in boron than any previously examined. The new procedure was used to produce the graphite for the first uranium-graphite pile, which was successfully operated in Chicago in December, 1942. Reference to the original problem and its solution in obtaining graphite of suitable purity is provided in Section 5 of Book IV, Volume 3, Manhattan District History. Paragraph 5-10a of that reference provides an incomplete statement in regard to the graphite suppliers. Early in the pile program interest was evidenced only by the Speer Carbon Company. That company provided a suitable product but did not possess the facilities for manufacture of the production pile moderator.

b. The Uranium Problem.-- Since metallic uranium had not been made hitherto in commercial quantities, it was necessary to develop methods for producing the metal, as well as to determine and control the purity of the materials used in its production. The investigation of both phases proceeded simultaneously. The preparation of uranium tetrachloride was begun by C. J. Rodden early in 1941. Soon thereafter fused pieces of metal were produced by the reduction of the chloride with calcium. In June, 1941, some of the pieces of metal thus obtained were remelted by J. G. Thompson. An important discovery resulting directly from this work was that the melting point of uranium was lower by at

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least 600° than the recorded value of approximately 1500° C. The boron content of this metal was too high, but it could be largely attributed to the calcium used for the reduction. Rodden and W. B. Holten found that if the calcium was first distilled before using it in the reduction process, satisfactory uranium was obtained. This work was interrupted in 1941, but in the early summer of 1942, the experiments on the reduction of uranium salts with distilled calcium were resumed. Excellent results were obtained when the tetrachloride was replaced by uranium tetrafluoride which had now become commercially available. Substitution of magnesium metal for calcium showed promise.

While the study of metal production was going forward, progress was also being made in the analyses of uranium compounds and in procedures for purifying them. Determinations of boron were now being made by B. F. Scribner by special spectrographic procedures that were standardized by means of samples chemically analysed by Scherrer.

During 1942, specifications for the purity of uranium compounds were prepared by a committee composed of Rodden, Swilard, R. Rosen, with F. K. Spedding as chairman. Methods of chemical analysis for many elements present as impurities were developed, and hundreds of samples were analyzed at the Bureau. For example, methods for the determination of rare earths in the impure oxide were of especial importance because of the known neutron absorbing properties of certain of the elements in this group. K. D. Fleischer and J. I. Hoffman contributed materially to the solution of this problem.

In conjunction with M. Pochon of the Eldorado Gold Mining Co., a method for obtaining purer black oxide was found which consisted

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of an acid leach of the material. The NBS contracted for the treatment of future shipments and the working over of some of the older shipments by this method.

As a result of the experimental work at the NBS on the purification of uranium oxide and on the distillation of calcium, it was evident in August of 1941 that raw materials for satisfactory uranium metal could be obtained or produced. However, at this time there was only one producer of metallic uranium, Metal Hydrides, Inc., at Beverly, Mass. The boron content of the powdered metal furnished by this company was so high (about 200 ppm) that it was practically useless for the purpose intended. C. J. Rodden, in an effort to improve the product, installed equipment at the Metal Hydrides plant for the distillation of calcium and showed that calcium hydride of high purity for reduction of uranium oxide could be made in large lots. In contrast with the metal first produced, with a boron content of approximately 200 ppm, the metal produced by Metal Hydrides after the Bureau's assistance contained less than 0.5 ppm of boron.

The decisive step in the purification of uranium compounds, prior to their conversion to chloride or fluoride and subsequent reduction to metal, was taken in the summer of 1941. At that time J. I. Hoffman showed that uranyl nitrate could be separated from all harmful elements by extraction with ether. By this method sufficiently pure uranium oxide could be obtained from crude uranium oxide and from pitchblende and carnotite ore concentrates. Ether extraction became the standard procedure for the final purification of all the uranium subsequently used in the piles.

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c. The Central Control Laboratory. When, in 1942, the Manhattan District assumed responsibility for procurement, the analytical work rapidly increased in scope and volume. Methods were now developed for the assay of uranium-bearing materials ranging from low grade carnotite to high grade pitchblende.

In February, 1943, when the Manhattan District set up a program for all raw and finished materials connected with the production of the metal, the NBS was designated as the Central Control Laboratory. As an illustration of the complexity of some of the work, one product required 17 individual chemical analyses in addition to the spectrographic determination of 29 elements. To aid other laboratories concerned with the metal control program, standard analyzed samples of some fifteen materials were prepared, including four kinds of uranium ore. Guest workers were instructed in analytical methods and other laboratories and plants operating under Manhattan District contracts were assisted in developing procedures and in overcoming difficulties.

Up to the end of 1945 nearly 9000 samples of materials had been received and nearly 30,000 separate analyses had been made. The developmental work on these problems of procurement resulted in contributions to 47 reports. In 1945, C. J. Rodden became chairman of the Manhattan District Analytical Committee.

The work of the Central Control Laboratory covered a number of phases of the project besides those relating to the procurement and analytical control of materials used in the piles. For example, in the latter part of 1941, C. J. Rodden and J. I. Hoffman developed a

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method for the purification and preparation of uranium compounds suitable for making films for determining isotopic composition by alpha-particle counting techniques. This was later put on a production basis and handled by L. J. Clark in cooperation with L. F. Curtiss, who was determining the degree of separation of U-235 obtained by the various investigators on isotopic concentration in the early work on uranium.

As further examples of the diverse nature of the problems that came to the central control laboratory, the following are of interest:

(1) In many cases uranium ores were purchased and paid for on the basis of their uranium content, with the provision that the radium be returned to the original owners in the residues left after removal of the uranium. Disagreements arose. To settle these, radium standards were prepared, in cooperation with L. F. Curtiss of the Radioactivity section for checking methods of determination of radium.

(2) Owing to the fact that the Metallurgical Laboratory at Chicago did not have adequate facilities for analytical control in their early work on plutonium, many materials involving special types of analyses were sent to the NBS. This included heavy water and the catalysts used in its production. Requests for assistance, especially in the capacity of consultants, continued through 1945.

(3) In cooperation with W. N. Harrison of the Clay and Silicate Products Division, ceramic materials were examined for suitability of their use in the electromagnetic process of isotope separation.

d. Spectrographic Analyses. The work of the Spectrochemical laboratory on the atomic bomb project was initiated in April 1940 when

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tests of the purity of several uranium oxide samples were made by qualitative spectrographic analysis. When the supervision of work on uranium was later assigned to the Office of Scientific Research and Development, the volume of testing increased and the samples then submitted included a variety of uranium compounds and other materials of importance to the project. It was apparent at that time that the usual spectrographic methods of analysis were inadequate for the analysis of uranium, especially in view of the increasing purity of the materials being produced.

The production of uranium of high purity was one of the serious problems encountered in the utilization of uranium as a source of atomic energy. Impurities, particularly the lighter elements such as boron, cadmium, and lithium, are effective absorbers of neutrons. Relatively small amounts of the impurities may reduce the efficiency of uranium fission below the point where a chain or self-sustaining nuclear reaction can be obtained. For many impurity elements, concentrations of a few parts per million were undesirable, and in the case of the most effective neutron absorbers, freedom from a few tenths of a part per million was essential. In order to control the production of pure uranium and to inspect the final product, methods of analysis were required for the determination of 60 or more chemical elements on a routine basis where speed, high sensitivity, and accuracy were important considerations.

The problems of the spectrographic analysis of uranium-base materials was complicated mainly by the highly-complex spectrum of uranium. Under the usual spectrographic procedures, interference by uranium lines would have rendered spectrographic analysis ineffective for impurity

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determinations at low concentrations. This obstacle was overcome by the development of four concentrational methods in which the impurities are separated from the uranium and then determined spectrographically.

The first satisfactory method for the spectrochemical analysis of uranium was developed at the National Bureau of Standards, and was described by B. F. Scribner and H. R. Mullin in April, 1942. It was physical in nature, involving fractional distillation of the impurities from an uranium oxide base. High sensitivity and accuracy were realized by adding a material, termed a "carrier", to the sample and distilling the carrier from the mixture by means of an electric arc. The carrier served to sweep the volatile impurities into the arc, the light from which was observed by a spectrograph. The high sensitivity of the method was attained by the almost complete suppression of the spectrum of uranium.

The carrier-distillation method was improved by a series of controls, including an electrode assembly of novel design. In June, 1942, the method provided for the determination of boron down to a limit of 0.1 part per million and of cadmium to 0.5 ppm, and included the determination of arsenic, cobalt, lithium, manganese, nickel, silver and sodium. The elements now determined by the method are Ag, Al, As, Au, B, Ba, Be, Bi, Cd, Co, Cr, Cs, Cu, Fe, Ge, Hg, In, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, Sb, Si, Sn, Tl, and Zn. The basic method was modified for the more accurate determination of iron, magnesium and manganese in February, 1945, thus obviating the need for chemical determinations of iron and manganese. The method provides for the rapid determination of the 32 elements at concentrations ranging from a few parts per million for most of the elements, down to one part in 100-million in the case of

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boron. The carrier-distillation method was adopted by other laboratories on the project and applied in the analysis of thousands of samples of uranium and similar materials. Over 120,000 determinations were made at the Bureau during the years 1943-45.

The three remaining methods that were devised for the analysis of uranium-base materials provided for the determination of the less-volatile impurity elements to which the carrier-distillation method did not apply. The spectrographic methods involved the analysis of three chemical concentrates from uranium-base samples. These concentrates, prepared by the chemical section under the supervision of G. J. Rodden, included the rare-earth group (Ce, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sc, Sm, Tb, Th, Tm, Y and Yb), the platinum-metal group (Ir, Pt, Rh and Ru) and a group of elements precipitated by cupferron (Co, Ga, Hf, Pd, Ta, Ti, V, W, and Zr). The required sensitivity of test was obtained by the combined chemical and spectrographic procedures and, in some cases, a remarkable limit was achieved. For example, the rare-earth method permitted the determination of the element ytterbium at a concentration of one part in 250-million parts of uranium.

The complete system of spectrographic methods for the analysis of uranium-base materials was recently described by Scribner and Mullin in the form of a 102-page laboratory manual. This manual, issued within the project in September 1945, provides detailed directions for the determination of 63 chemical elements in 11 types of uranium-base materials. The spectrographic system, combined with extensive chemical analytical work, distinguishes uranium as the most thoroughly analyzed

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material in the history of analytical chemistry.

e. Metallurgical Investigation of Uranium. Phases of the metallurgy of uranium investigated at the Bureau included the development of methods for melting and casting the metal, methods for working uranium, and investigation of the uranium-oxygen system, which necessarily included development of methods for determining oxygen in uranium and for metallographic examination.

Investigations on thorium included a few experiments on comparison of beryllia and thoria as melting refractories for the metal, determination of density and hardness, development of procedures for preparing metallographic specimens, and study of the microstructure of cast and extruded thorium.

(1) Melting and casting uranium: In the early stages of the project, utilization of uranium required the development of methods for melting and casting to permit producing the metal in the desired shapes, and also to eliminate, in part at least, certain deleterious impurities.

Investigation of the melting of uranium was initiated at the Bureau in May 1941, using metal prepared by C. J. Rodden. Work on the problem was sporadic until April 1942 when metal from contract producers became available and extensive research on the metallurgy of uranium began. The technique of casting progressed from the production of 100-gram ingots to melting approximately 50 pounds of uranium at a time. Contributions of the Metallurgical Division to the technique of melting uranium included: (a) identification of beryllia as the first material in which uranium could be melted successfully; (b) demonstration that metal molds, as well as graphite molds, could be used for casting uranium;

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and (c) development of a procedure for casting bars from each of which two or more 5-inch slugs for the pile could be fabricated. The cast bars were superior to extruded rod, in regard to interior defects, and adoption of the casting process avoided the difficult extrusion process. Nearly three tons of uranium were cast in bar form at the Bureau.

(2) Development of methods for fabricating uranium: The hot and cold working properties of uranium were investigated and procedures for forging, rolling, swaging and wire drawing were developed. Forging was performed most satisfactorily at temperatures just below the alpha-beta transformation (665° to 670° C). The metal was more plastic in the gamma state (above 760° to 770° C) and deformed more readily, but if cooling into the beta range occurred during forging, fracture of the hard brittle beta phase resulted.

Hot rolling, at temperatures near the top of the alpha range, was far more rapid than cold rolling, but the surface obtained was inferior because of the amount of oxide rolled in. Cold rolling with frequent anneals was a slower but more satisfactory procedure. For the same reason cold swaging was preferable to hot swaging. An additional difficulty experienced in hot swaging was cracking of the metal as it transformed into the brittle beta phase, as one result of overheating in the die.

Bare uranium could not be drawn satisfactorily with any of the oil or grease lubricants customarily used in drawing other metals. Use of copper as a lubricant, by electrodepositing copper on the cleaned wire, prevented jamming of the wire in the dies and permitted drawing wire to 10 mils diameter.

Anodic cleaning of the wire was a necessary preliminary to plating.

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When the anodic cleaning and the plating were conducted in different baths, film formation during exposure to the air while transferring from the cleaning to the plating bath frequently prevented satisfactory adhesion of the plating. A procedure was developed for cleaning anodically and plating in the same bath, avoiding exposure of the wire to the air and resulting in a satisfactorily adherent plating. After drawing to the desired diameter, pickling in a 60 HNO₃-40 H₂O mixture removed the copper coating.

(3) Other metallurgical research included: the determination of the solubility of oxygen in molten and solid uranium; determination of oxygen in uranium by vacuum fusion; oxidation of uranium in air at temperatures up to 500° C; bright annealing of uranium; and metallography of uranium.

f. Physical and Mechanical Properties of Uranium. Much of the information in the literature on the properties of uranium was inaccurate because it represented the properties of the impure metal previously available and not those of the relatively pure metal produced by methods developed recently.

Temperatures for the melting point of uranium given in the literature vary from 1300 to 1850° C, the latter temperature being given in most handbooks. Early melting experiments at the Bureau showed the melting point to be much lower than the reported values. The metal available in 1942 melted in the range 1075 to 1125° C. Precise melting point determinations were later made by the Heat and Power Division on four specimens of uranium produced late in 1943. One was melted in beryllia, one in thoria, and two in graphite. The best value for the

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melting point of uranium, with 5 parts per million of carbon, 60 parts per million of iron, 20 parts per million of oxygen, and lesser amounts of other impurities, was $1132 \pm 1^\circ \text{C}$.

The other properties determined were: density; electrical resistivity and temperature coefficient of resistance; thermo-electric potential; specific heat of uranium and its compounds at high temperatures; transformation points; hardness; tensile properties; compressive properties; and physico-chemical data on uranium compounds.

g. *The First Spectrum of Uranium.* At the beginning of the year 1942 there were only fragmentary descriptions of the spectra emitted by neutral and ionized uranium atoms. No Zeeman effects had been observed; no term analyses had been made; nothing was known of the configuration of the valence electrons; and no value had been found for the nuclear moment of the odd isotopes. It was known, however, that the emission spectrum observed in the ordinary arc and spark sources is exceedingly complex, and does not at all resemble the spectra of chromium, molybdenum, and tungsten, which precede it in column VI of the periodic table of the chemical elements.

A much clearer understanding of the structure of the uranium atom was necessary in connection with other investigations that were under way, and the Spectroscopy section was requested to undertake an investigation of the uranium spectra.

The program of work comprised the observation of the uranium spectra as emitted by arcs and spark discharges in air at normal and at reduced pressures, by discharges in magnetic fields, in hollow cathodes, and by under-water spark discharges. Available for making

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the observations were four concave-grating spectrographs and a large quartz-prism spectrograph, together with auxiliary apparatus such as Fabry-Perot interferometers and Lummer-Gehrcke plates.

A separation was effected of the lines emitted by neutral uranium atoms from those emitted by ions. A list of more than 9,000 lines with accurate wave lengths, wave-numbers, and estimated intensities, for the region 3,000A-11, 000A, were compiled as descriptive of the spectrum of the neutral atom. A partial analysis of this spectrum was made by the classification of about 2,000 lines as combinations between 18 low and metastable odd energy levels with more than 280 higher even levels. On the basis of well-resolved Zeeman effects several of the low levels were identified as components of $5L$, $5K$, $7M$, and $7K$ terms arising in the electron configurations f^3d^2 and f^3d^2s . The spectrum of uranium was interpreted as that of a rare-earth element analogous to that of neodymium, uranium being designated as the third member of a second group of rare-earths beginning with thorium. Easy ionization in the electric arc and in magnetic fields, and the short-wave limit of the observed spectrum indicated that the ionization potential of uranium atoms is approximately four volts.

h. Concentration of Isotopes. Two of the miscellaneous investigations of the process of uranium isotopic separation originated at the National Bureau of Standards. These were the electromigration and the molecular distillation methods. While some degree of separation was attained by the latter method, nevertheless, both methods proved in the main to be unsatisfactory for the purpose intended. General accounts of the National Bureau of Standards and other organizational activities for the electromigration and the molecular distillation separation methods

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are provided in Book I, Volume 4, Chapter 14, of the Manhattan District History.

1. Mass Spectrometer Analyses. At the outset of the war, the National Bureau of Standards did not have a mass spectrometer laboratory. When the electrolytic migration method for the separation of potassium isotopes was devised, it was necessary to set up at the Bureau the mass spectrometer which had been built at the Fixed Nitrogen Research Laboratory by A. K. Brewer to make the analyses. This marked the beginning of the present Mass Spectrometer laboratory. During the time that the electromigration method was being investigated, the mass spectrometer was run about 16 hours per day.

After successful methods had been worked out for the concentration of potassium isotopes, the methods were applied to uranium. Since uranium isotopes could not be analysed on the potassium mass spectrometer, it was necessary to obtain an instrument for this purpose. Fortunately, the Bureau was able to acquire the parts of a mass spectrometer built by Dr. O. A. Nier at the University of Minnesota.

The mass spectrometer was designed to analyse uranium in the form of gaseous UF_6 . This gas upon ionization by electron impact in the mass spectrometer is dissociated into all possible combinations of uranium and fluorine, UF_5 being the most abundant. In consequence, the 330 and 333 peaks were used in determining the isotope ratios. The original method of operation consisted of reading the two mass peaks simultaneously, the big 333 peak being read on a microammeter and the small 330 peak on a galvanometer. In order to enhance the sensitivity of the instrument, the entire electrical pick-up for the 330 peak was replaced with a more

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stable design while the circuit for the 333 peak was revised and re-insulated. While these changes materially enhanced the stability and sensitivity of the instrument, the actual recording of data was still cumbersome since it was impossible for one observer to read the deflections for both peaks at the same time. In addition to reading the meters, the operator was required to operate the voltage scanning dials by hand to bring into focus the mass peaks.

In order to simplify the operation of the mass spectrometer as well as to increase the accuracy of observation, an automatic voltage sweep and recording mechanism were designed. The recorder was assembled from parts of commercial Brown instruments. The scanning circuit was a modification of that used on the Consolidated mass spectrometer, in that the accelerating voltage was changed by allowing the potential on a condenser to decay through a high resistance. When the potential had changed over the required range, a relay arrangement allowed the condenser to be recharged and the process repeated. In order to eliminate all manual recording of data during a run, a control was placed in the ionizing circuit which permitted the operator to keep the 333 isotope beam as a constant value. This recorder not only vastly simplified the operation of the mass spectrometer but also increased the precision and shortened the time of analysis. It was later widely used at Oak Ridge in recording the progressive changes in concentration in the great production plants.

J. Radioactivity Measurements. In the early part of the development of the atomic bomb it was necessary to have a method of testing various procedures for separating the isotopes of uranium. In particular, it was desirable to be able to make measurements which would

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reveal to what extent the percentage of uranium 235 had been increased in a sample by the particular treatment in question, and to detect small increases in this percentage. This permitted the efficacy of various proposals for this isotopic enrichment to be tested at the early stages and thus permit a selection of the best, with a minimum loss of time.

A method was developed in the Radioactivity section, based on the rate of emission of alpha particles from the sample of uranium, using the alpha particles from the uranium 234 isotope as an indicator. Since the percentage of enrichment for the 235 and 234 isotopes depend on their masses relative to the mass of the third, the 238 isotope, any treatment which enriches 235 will also increase the amount of 234 in a simple ratio of the masses. However, in the natural sample there are just as many alpha particles from 238 as from 234 because the 234 is produced from 238 and the process has been going on in nature a sufficient time for the amounts of each to reach a steady value. If the relative amount of 234 is increased, and with it the relative amount of 235, there will be more alpha particles per gram per second from the enriched sample than from a natural untreated sample. This method was made sufficiently accurate to detect a change of 1/10 percent in the enrichment. To do this at least 1,000,000 alpha particles must be counted reliably and accurately from each sample.

Equipment to do this work was developed and constructed in the radioactivity section and numerous samples were tested for various laboratories, which aided greatly in selecting suitable methods of enrichment. In particular, the measurements made at the NBS were very helpful to Dr. Abelson of the Naval Research Laboratory in the develop-

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ment of his process for isotopic separation.

As a by-product of the work in counting alpha particles from uranium, the radioactivity laboratory prepared many carefully weighed samples of uranium deposited in accurately formed disks on platinum. These samples constitute reliable sources of beta-rays which can be used as standards in the calibration of beta-ray counters. Sets of these standards were furnished various laboratories engaged in radioactive work in connection with the war.

In connection with radioactive war-work, it became desirable to have many Geiger-Muller counters available which had the thinnest possible aluminum walls, so that they could be used to measure beta-rays. Previous methods of producing such counters were slow and expensive. The radioactivity section developed a method of copper plating thin aluminum toothpaste tubes so that fittings could be attached to them by ordinary soft solder to convert them into Geiger-Muller tube counters. The operation was very simple and cheap so that several counters could be made in the time previously required to make one, with a corresponding reduction in cost. These toothpaste tubes also had the minimum thickness required for evacuation and filling without collapsing the tube.

For the identification and estimation of radioactive isotopes which emit alpha rays, it is desirable to have a counting equipment which will separate the particles of the same range characteristic of each isotope and count them separately. An electronic circuit was developed which would register the pulse produced by the amplification of the ionization resulting from a single alpha particle of given range. All other pulses were rejected by this particular circuit. By arranging a sufficient

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number of these circuits, adjusted so that they just covered the various sizes of pulses to be anticipated, the actual distribution of the alpha particles in terms of their ranges from a particular source could be found. It proved to be of great value in identifying and measuring isotopic mixtures of alpha-ray emitters when the number of particles from a particular isotopes exceeded 2 or 3% of the total number. It was used to check the percentage and range of the alpha particles from U235 in natural uranium and in an unsuccessful search for U236 as an alpha-ray emitter in irradiated uranium.

One of the problems in the procurement of uranium ore was the determination of the quantity of radium in the ore as well as the quantity of uranium, for both these elements determine the value of the ore. This problem was solved by utilizing the technique of radon measurement, developed for another purpose, in which alpha particles are counted.

During the war, plants were established by the Army and Navy for applying radium luminous compounds to dials of instruments. The application of such compounds can result in serious injury to personnel unless proper regulations and tests are established. The measurement of radon in the exhaled breath of radium dial painters was thus mandatory. The radioactivity section of the Bureau was instrumental in the development of suitable regulations and made the tests of expired air to determine the efficacy of these regulations and their enforcement. In addition to the apparatus for determining the radon content of samples of air the NBS designed a glass bulb for shipping samples of air which could be sent evacuated to the collecting point to facilitate the transfer of the sample to the flask. This service for the Army and Navy was

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of considerable value to them, since without it they would have hesitated to engage civilians to apply radium luminous compounds and expose them to the well known dangers of radium poisoning. As a result the hazards have been reduced to a few cases of temporary over-exposure.

The procurement of large quantities of uranium ore by the Government required that the radium in the ore be determined as well as the uranium, in arriving at a value for the ore. For example, a ton of high grade uranium ore might contain as much as \$2,000 worth of radium. In addition, the measurement of the radium gives an indirect check on the amount of uranium since there is a definite ratio between the radium and uranium in natural ores.

The same general equipment used for measuring breath samples was also used to measure radium in uranium ores. The chief difference in the procedure is that, for measurement of radium, the ore sample is dissolved and the radon accumulated in the solution over a definite time. The radon is then transferred to the counting chamber where the alpha particles are counted. From this procedure the amount of radon is determined. From the amount of radon and the time during which it was accumulated the amount of radium in the sample can be computed.

Several thousand such measurements were made. As a part of the program, comparative measurements were made with other laboratories which carried out determinations for various contractors. As a result it was found that the values obtained by the Bureau were both more consistent within themselves and nearer the average values of all other laboratories. This resulted in the selection of the Bureau as a referee in case of disagreement of measurements made elsewhere.

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