History and accomplishments of the

OAK RIDGE GRAPHITE REACTOR

By Arthur H. Snell and Alvin M. Weinberg

At 4:00 A.M. on November 4, 1943, Louis Slotin knocked on the doors of the Oak Ridge houses of M. D. Whitaker and R. L. Doan, directors of the Clinton Laboratories of the Manhattan District. Through the night, uranium slugs had been continuously loaded into the closely-guarded Graphite Reactor at "Site X" in the rolling hills of East Tennessee, and Whitaker and Doan had left strict instructions that criticality should not be achieved until after they had arrived at work the following morning. However, the critical mass had been overestimated, and the enthusiasm of Henry Newson, Lyle Borst, and Slotin had perhaps been underestimated; at any rate, criticality came sooner than expected, and Slotin found it necessary to jump into a car, drive into town, and rouse the bosses from their beds for a dusty but starlit drive of fifteen miles to the reactor site.

On November 4, 1963, Dr. Doan pressed a button that closed down the Oak Ridge graphite reactor after twenty years of continuous operation (Fig. 1). The occasion was marked by a reunion, and although death had taken Slotin and Whitaker, the occasion was one of reminiscence. Those who helped to design and construct the "Clinton pile" had given little thought to the possibility that this war-time "intermediate plant" would become an instrument of extraordinary power and versatility, or that it would be a nucleus from which a flourishing laboratory would grow. Yet during those twenty years the graphite reactor was undeniably one of the world's most important scientific instruments; it spawned new science and new technology to a degree not remotely suspected earlier in 1943, when the pile was being planned. It was an appropriate occasion, therefore, to review the accomplishment of this historic reactor in several fields of technology and basic science.

Reactor technology

Within a few days of the attainment of the fission chain reaction, it was realized that an air-cooled pile could be rather readily built, and this was the genesis of the graphite reactor. The famous experiment of Fermi and his associates in Chicago took place in an unshielded assembly, in the heart of a great city, and CP-1 could be safely flashed to powers of one or two kilowatts on one or two occasions only. The Clinton pile was shielded and cooled for 1000 kW operation, and was remotely located. Inasmuch as it was air-cooled, it was not a pilot plant for water-cooled Hanford, but it was truly a pilot plant in showing that a nuclear reactor could be continuously operated at full power and temperature, with complete safety. It also pioneered the rod lattice, in contrast to the cubic lattice of CP-1. The confidence that it engendered in the whole Plutonium Project was of course enormous. Its other mission was to produce a few grams of plutonium, to feed to an adjacent pilot plant which modelled the chemical separation plants in the great "canyons" of Hanford. This mission it accomplished with complete success.

The necessity for extracting sizable amounts of heat from the uranium obliged the designers of the graphite reactor to face up, for the first time, to one of the central problems of reactor technology: fabrication and canning of fuel elements. The experience, sometimes frustrating, of making an acceptable aluminum-clad, metallic-uranium slug was invaluable to those who had to clad slugs for Hanford. That canning a uranium slug with

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aluminum could be devilishly tricky—almost proving the undoing of the entire Hanford project—really dawned when attempts were made to can these slugs. Even to decide whether a slug was canned properly was not easy—Lyle Borst, who was then in charge of testing the slugs, devised a method whereby they were individually accurately weighed before and after a ten-day heating period; an increase in weight implied oxidation, and therefore a faulty seal in the jacket. Laborious the method certainly was (a loading required some 43 000 slugs!), but it was adequate and therefore welcome under the stress of the time.

The nuclear design of the Clinton Reactor presented few problems—it was done as sort of a part-time effort, a diversion from the much more exacting job of designing the Hanford lattice. Two points are worth recalling: first, the theory of the control rod was really originated for the graphite reactor; second, the two-group theory—forerunner of the modern complicated *n*-group machine calculations—was used for the first time to estimate the reflector savings of a reactor.*

The graphite reactor initially missed the matter of ^{135}Xe poisoning. Of course nobody knew that the ^{135}Xe fission product had so large a cross section at the time the reactor was designed—it was probably for the better since the project in those days could hardly tolerate an additional one-half percent or so reduction in the multiplication constant. With the reactor operating at 500 kW the loss due to Xe was around one fortieth of a percent—enough to observe, as subsequent observations showed. Yet the Xe poisoning remained unnoticed until after it showed up with almost devastating consequences at Hanford. Looking back, we surmise that the reason was that the reactor had a negative temperature coefficient; hence, as the reactor warmed up after reaching power, the rods had to be pulled out as a matter of routine. Unless one were on the lookout for a secular loss of reactivity that saturated after about 20 hours, it would have been most unlikely to see the Xe effect which at 500 kW, amounted to 11 inhours, or to moving the shim rods 15 inches in 20 hours. Borst and others later saw it very clearly, when they knew what to look for.

Fig. 1. The shut-down ceremony for the Oak Ridge graphite reactor. The speaker is R. L. Doan.

Although the xenon effect was not discovered in the graphite reactor, it was extensively measured in some later nuclear measurements. Two "negative activation" experiments were performed by teams in the Chemistry and Physics Divisions, which gave the ^{135}Xe cross section in sufficient accuracy for pile neutrons. Another experiment, performed by Seymour Bernstein, Maurice Shapiro, George Parker, and others, was a tour de force (for that time) in which 10-curie samples of fission product ^{135}I and Pd^{135}I were placed in the neutron beam of a crystal spectrometer, and the growth of the capture-cross-section peak of the daughter ^{135}Xe was measured in detail over the energy range 0.01 to 0.2 eV. Then later still, just to show there was no fooling, the energy range was extended by using 500-curie samples of ^{135}Xe from the homogeneous reactor. This was done by Pawlicki, Smith, and Thurlow with a rotating chopper, but it did not involve the graphite reactor.

The graphite reactor also contributed directly to the newly developing reactor physics. Two major experiments were performed during 1944. The first measured the relaxation length of neutrons in a large pile of natural uranium. From the relaxation length, one could determine E, the number of neutrons produced by fast fission in natural

* In this connection, one recalls an amusing incident concerning Fermi, who of course was a one-man theoretical department. He always calculated everything he needed himself; the young theoreticians would try only to get answers that agreed with Fermi's private calculations. The design of the Graphite Reactor lattice was no exception—Fermi had his own private calculations of the whole reactor. Included was a calculation of the reflector saving, but since the group method had not received much attention at that time, Fermi used the far more elaborate age theory (which amounts to using an infinite number of groups). Fermi described his calculations at a theoretical colloquium: he came out with the surprising result that the reflector saving according to age theory was less than that calculated according to the simplest, one-group theory then in vogue. Several young theoretikers had made the same calculation according to two-group theory, but obtained the opposite result—that the true reflector saving exceeded the value given by one-group theory. But Fermi's explanation was persuasive, and the best the theoretical group could do was to urge him into recalculating his result by two-group theory. This he did, and he never again suggested that the true reflector saving was less than that calculated by one-group theory.

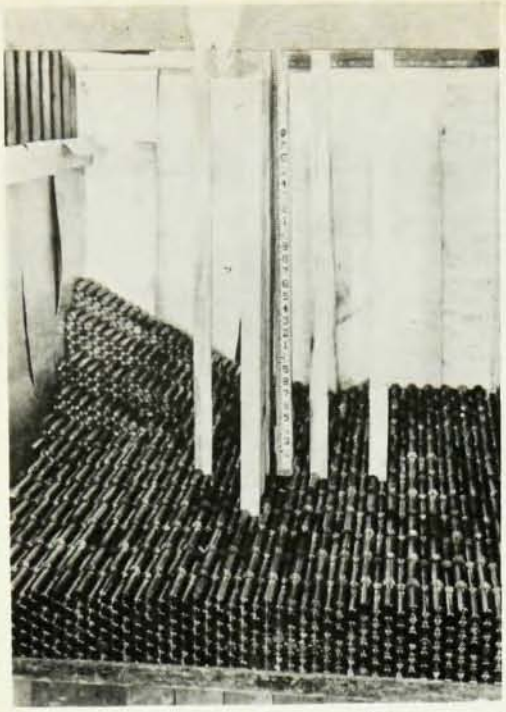

Fig. 2. The uranium-metal exponential experiment repeated at Clinton in 1944. Photograph shows a partial loading of the stack of uranium-metal slugs in the thermal-column aperture in the top shield of the graphite reactor. The vertical metal channels were slots for foils. Below the slugs there was a layer of graphite and below that the active lattice of the reactor.

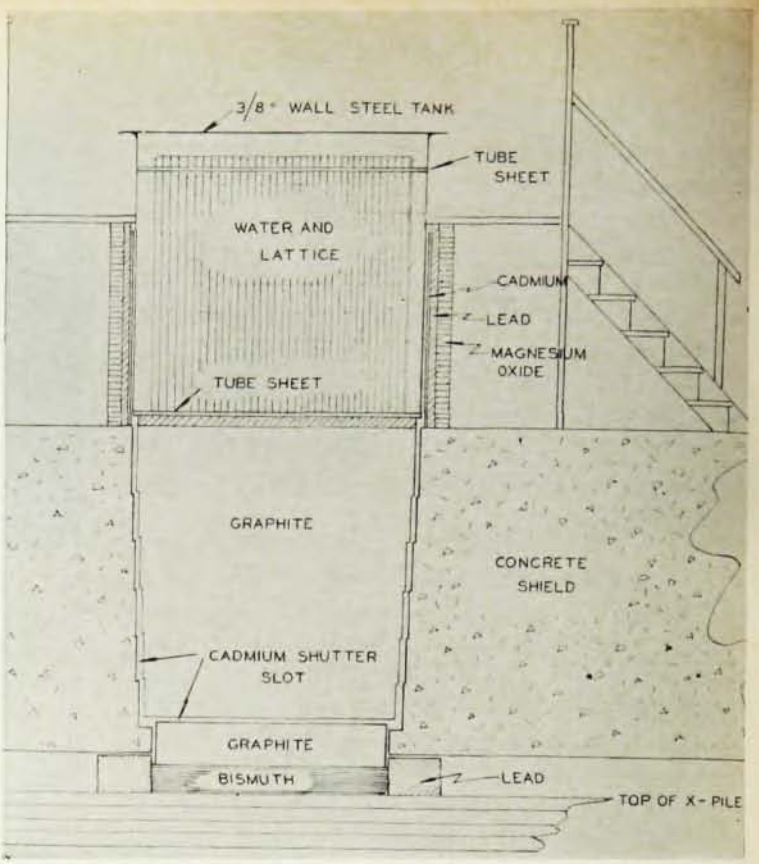

Fig. 3. Cross section of the water-lattice experiment which yielded a reproduction factor of very nearly unity with natural uranium and natural water.

uranium per neutron produced by slow fission in ^{235}U. If this number exceeded one, a fast-neutron chain reaction in natural uranium would have been possible. Such a possibility could not be ruled out in the earliest days of the Metallurgical Project, and one of us was assigned the task, at the Chicago cyclotron, of answering this crucial question. The first experiment at Chicago used the first five tons of uranium metal that had been produced, and gave the reassuringly low value $E = 0.2$;* but because the block of uranium was so small, and the corrections for neutrons leaking out so large, the project decided to repeat the experiment at Clinton with 35 tons of U (Fig. 2). [The slugs used in the pile (erected on top of the graphite reactor) were stolen temporarily from those eventually destined for the graphite reactor itself.] The experiments again gave the value $E = 0.2$. Clearly neither the Germans (nor we, for that matter) could make an atomic bomb by piling up enough natural uranium; one had to face the long, tough road of enrichment or plutonium production.

* This meant that uranium would have to be enriched by a factor of 11 in order to sustain a fast-neutron chain reaction.

At about the same time, it became theoretically apparent that the addition of ordinary water might bring the assembly very close to criticality. The change to the water exponential experiments was an easy step—the same thermal column was used; almost all that had to be done was to reload the tank with vertical rods, and add water (Fig. 3). The experiments were performed by a large group that included Louis Slotin, Henry Newson, Haydn Jones, and Seymour Bernstein. The infinite-size multiplication constants found in some of these experiments were extremely close to one. Much later experiments at Brookhaven by Kouts have verified what at that time seemed to be an astonishing result: that an unenriched-uranium–ordinary-water lattice was just about chain reacting. The very high multiplication constant in these lattices was attributed to the strong "interaction" fast effect: fast neutrons from one rod could, with high probability, traverse the water without losing energy and induce fissions in neighboring rods. This interaction fast effect adds about eight percent to the fast effect in some of the more closely packed H_2O-U lattices.

That natural uranium and water could probably sustain a chain reaction was less important than we first believed since at the time we knew nothing

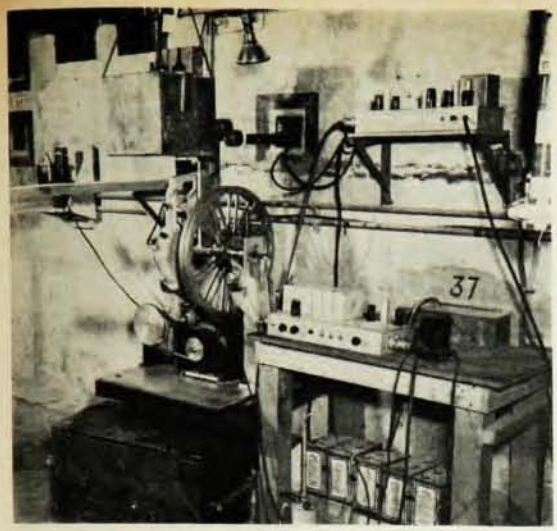

Fig. 4. Early pile oscillator.

about the ^{135}Xe poisoning. Nevertheless the early experiments at what is now Oak Ridge certainly helped focus attention on the possibilities of using pressurized water in a power plant. Many of the experimental and theoretical techniques devised in the course of the earliest experiments on light-water lattices at Oak Ridge are still used today in the design of many slightly enriched uranium pressurized or boiling-water power reactors.

Any mention of the technological work done on the graphite reactor would be incomplete without mention of the early Hanford shield tests, and the many shielding experiments subsequently performed with the Lid Tank. Out of these activities came the pool reactor, the tower reactor, naval reactor shields, and now even work on space-vehicle shields!

Another development, on the borderline between reactor technology and physics, was the pile oscillator. The idea was due to Eugene Wigner, the execution was by Ernest Wollan, Walter Jordan, and others, and the measurements were mostly by Herbert Pomerance. Figure 4 shows the Maytag washing machine that Wollan adapted for pulling strings so as to give a reciprocating motion to the sample whose poisoning effect on the pile was to be measured. An important result of this work was the discovery of the low neutron-capture cross section of zirconium, with the consequence that zirconium has had an important place in reactor technology ever since.

Radioisotopes

History may well assign to the graphite reactor the honor of making the radioisotope a major scientific tool. The progress that is being made in working out the details of protein synthesis, in our whole revolutionary insight into the basic life processes, has been possible only because ^{14}C, tritium, and ^{32}P are now everyday laboratory chemicals. Before the graphite reactor this was not the case; after the graphite reactor, radioisotopes became the central technique in the most ramified branches of science and technology.

This change did not happen automatically. It was largely the persistence immediately after the war of Waldo Cohn and Paul Aebersold that got the radioisotope business going in a major way and the continued devotion of Arthur Rupp that kept it flourishing. There were problems of administration and problems of science. The first 440 millicuries of ^{14}C were produced by an experimental method (Fig. 5) in which ammonium-nitrate solution was circulated through the pile and the ^{14}C came off as $^{14}CO_2$ or ^{14}CO, to be precipitated out by bubbling through barium-hydroxide solution. Some of the product was brought to 40 percent isotopic purity by Dewey Norris, and it was a long time before such a high specific activity was again attained after the process was changed

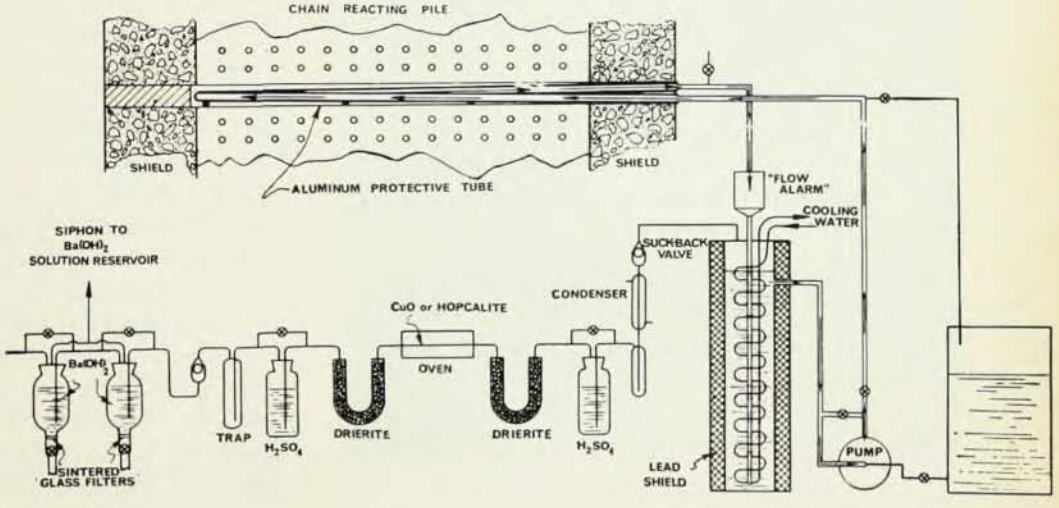

Fig. 5. Early production of carbon 14. A pump circulated an ammonium-nitrate solution through the reactor. At the top of the heat exchanger, off-gases were dried and prepared for precipitation of ^{14}C and $Ba^{14}CO_3$ (far left).

to the chemically more sophisticated Be_3N_2 method. The production of ^{32}P by the sulphur (n,p) reaction had been a laboratory curiosity until it was performed on a large scale at the graphite reactor; and also the production of T by Li (n,α) by Vernon Cannon, Norman Elliott, Glenn Jenks, and Edward Shapiro, and the measurement of its optical spectrum by Pomerance; and ^{131}I from fission products. All these may seem trivial and simple compared to our modern production of hundreds of thousands of curies of strontium 90, or megacuries of cobalt 60, but at the time, they were revolutionary, and difficult. It may be no exaggeration to say that the contribution of the graphite reactor to the technology of isotope production will stand as its most important achievement—in the annals of mankind, probably more important than its original production of plutonium. All who had even a small part to play in this achievement must take great pride in the profound influence that the work has had on all branches of science and technology.

Physics

In the contributions of the graphite reactor to physics, we believe that neutron diffraction must stand high on the list of importance because of the place that it has since established for itself in solid-state physics and in structural chemistry. A little work on neutron diffraction was done at the CP-3 pile at Argonne in the early days, but the subject really got going when Wollan brought from Chicago an old x-ray goniometer and set it up in his first neutron diffractometer shown in Fig. 6.

As one looks at neutron diffraction as developed

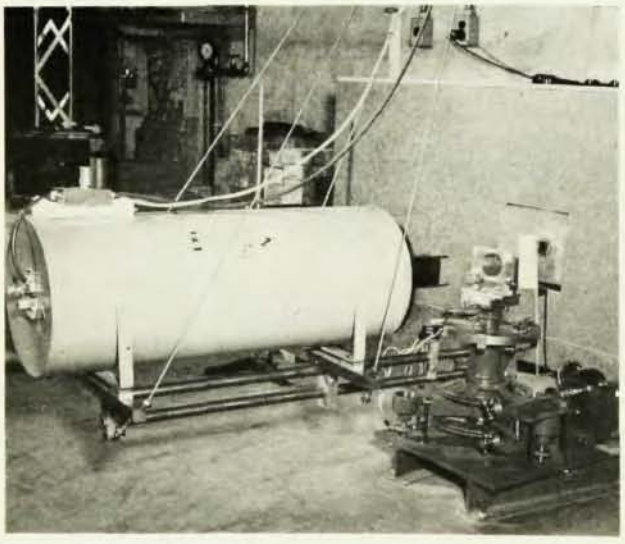

Fig. 6. Early neutron-diffraction apparatus (Wollan)

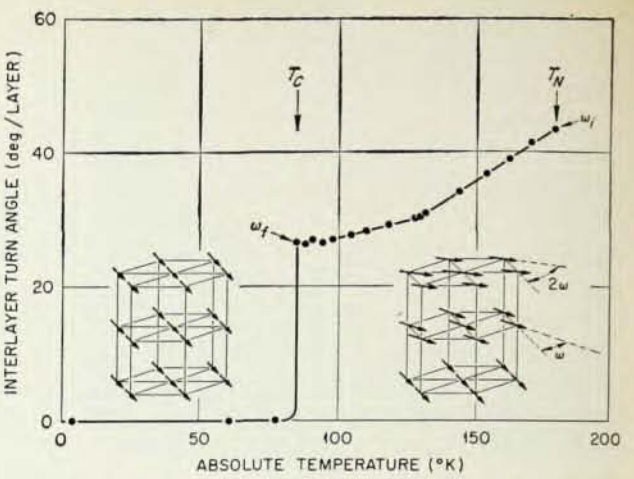

Fig. 7. Temperature dependence of structure in dysprosium from magnetic-structure studies by neutron diffraction developed by E. O. Wollan and his associates

here, three main courses are evident. The first is in nuclear physics, where Wollan and his associates measured neutron coherent scattering amplitudes for most of the elements, and where the neutron-proton interaction had special importance. The second is in the study of the magnetic structure of solids where, as a result of the leadership of Wollan and Shull, a whole science has developed in the study of magnetic lattices and their transitions from one kind to another as the result of changes of temperature, composition, or the direction of an external magnetic field. Figure 7, for example, shows a twisting of the magnetic structure in dysprosium that sets in above 50°K, and increases with temperature. All of this of course is possible because the neutron has a magnetic moment, and scatters coherently from these beautiful magnetic superlattices.

The work on the location of hydrogen atoms was picked up by Henri Levy, Selmer Peterson, and others. Here the neutrons have the advantage that they are scattered by hydrogen about as much as they are scattered by other elements, so hydrogen atoms are revealed in substances in which they are invisible to x rays. Here again there is obviously a whole science, broader even than the magnetic work. By way of example, Fig. 8 shows a two-dimensional display of the structure of sucrose as worked out by Levy and his colleagues.

A second notable success at the graphite reactor has been the work of Louis Roberts, Seymour Bernstein, and their collaborators on nuclear alignment, again a field where solid-state and nuclear physics meet. In 1948, when Roberts began his work, the possibility of polarizing nuclei seemed remote. What was needed was a magnetic field of around

50 000 oersteds at 0.1°K. The outlook brightened when M. E. Rose and C. J. Gorter independently suggested (in 1948) that the magnetic field at the nucleus due to unpaired electrons in the transition elements might be 10^6 oersteds or more: hence, if one polarized the electronic moments, the nuclei in such elements would obligingly line up also. The experiment was first performed by Roberts and Bernstein on Mn—the advantage of doing it at the reactor was that the reactor provided a convenient and intense beam of polarized neutrons. These could be used to test in a unique and highly sensitive way how many of the nuclei had been polarized. During the subsequent 14 years that such experiments have been performed, a dozen nuclei have been aligned by the Rose-Gorter technique, and the spins of about the same number of compound nuclear states determined in an absolute manner. Perhaps more significantly, the courage displayed by Roberts and Bernstein in embarking on so doubtful and difficult a project, and the success they achieved (added to the success of a strong group at Oxford), encouraged many others to think about nuclear alignment. It had been brought within reach. Roberts and Dabbs went on to perform experiments on the fission of oriented nuclei at the graphite reactor. Nuclei have now been aligned by several different techniques, of which the Rose-Gorter method, the brute-force method, and the electric quadrupole coupling method have been exploited at Oak Ridge.

Several experiments at the graphite reactor were concerned with the properties of the neutron as an elementary particle. First was the direct demonstration of the radioactive decay of the neutron. Though the neutron intensities available at the reactor were too low to permit a measurement of the half-life with precision, these early experiments influenced later workers who had higher intensities available to them. Out of these experiments eventually grew the highly sophisticated experiments on the β-decay of ^{6}He, and the field of charge spectrometry.

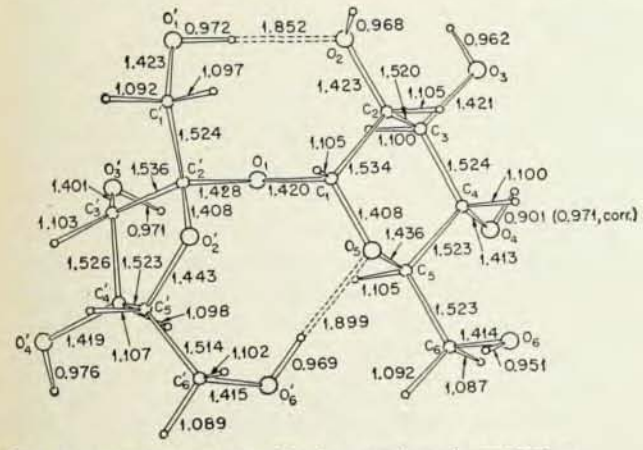

Fig. 8. Accurate location of hydrogen atoms in sucrose—part of a program of crystal-structure studies by H. A. Levy and his associates

The elementary properties of the neutron were also involved in two other experiments: a Stern-Gerlach experiment on polarized neutrons performed by Sherwood, Bernstein, and Stevenson, and a sensitive experiment by James Smith and Norman Ramsey which showed that the neutron does not have an electric dipole moment greater than 5×10^{-20} e-cm, where e is the electronic charge.

Solid state

Immediately after the war the graphite reactor became the site of many important experiments on the effect of irradiation on matter. Perhaps some of the most significant were the early observations, instigated by Sidney Siegel, Woody Johnson, and Karl Lark-Horowitz, of the effect of neutron irradiation on semiconductors. The striking sensitivity of semiconductors to heavy-particle radiation had not been fully appreciated until Lark-Horowitz's monthly visits to Oak Ridge. These experiments proved to be particularly important after the transistor was invented: the balkiness of transistorized devices in radiation fields was foreshadowed by the work at Oak Ridge on irradiation of semiconductors.

Research in radiation damage at the graphite reactor has traditionally been concerned with structural metals. It began with the early experiments of Billington and Siegel, who studied the effect of radiation on the strength, hardness, and elasticity of structural metals, and on the resistivity and hardness of order-disorder and precipitation-hardened alloys. Similar studies are continuing even today with the elaborate technique of irradiating materials at low temperature—first at liquid-nitrogen, then down to liquid-hydrogen temperatures, and now at 3°K. The cryostat in the graphite reactor, designed and built by Thomas Blewitt and Ralph Coltman, was the first such low-temperature facility in an operating reactor. The facility was a by-product of the original hydrogen bomb tests—the huge hydrogen liquifiers, no longer needed at Eniwetok, diffused to Oak Ridge where they became part of the Blewitt-Coltman low-temperature facility. The apparatus made it possible for the first time to measure "in-pile" electrical resistivity, stored energy, critical shear stress, dimensional stability, and internal friction at liquid-helium temperature. In regard to the internal-friction

experiment, a novel scheme for transmitting elastic data was devised by D. O. Thompson; viz., an in-pile radio transmitter.

The graphite reactor has been used by solid-state people long after most nuclear physicists and chemists have moved to newer and more intense sources. The stability of the flux, the low level of gamma heating per unit of flux, and the large volume of the reactor have so endeared it to them that they were the last scientific tenants to be evicted from it.

Nuclear and radiation chemistry

From the beginning, chemists turned to the graphite reactor as a source of new elements and isotopes. The production of Pu needs no more comment. Promethium was discovered here by Glendenin and Marinsky, and it was here that the first weighable amounts of technetium were produced.

The characterization of new isotopes has been a major activity of radiochemists from the early days when Charles Coryell and his associates filled out the classic fission yield curve until the present day of scintillators, solid-state detectors and 20 000-channel analyzers. Most of the activities were produced by the (n,γ) and $(n,\text{fission})$ reactions, but one clever variant made use of the tritons from $^{6}\text{Li}\,(n,\text{T})\,^{4}\text{He}$ to produce secondary reactions: a few positron emitters were even made in this way. It would be difficult to speculate on how many thousands of irradiations have been made in the graphite reactor, but one *can* say that most have been done in the pneumatic tube. That utilitarian gadget was installed by the chemists in the earliest days, and has proved so indispensable that it is almost part of the pile, and faster versions have been developed for work on short-lived radioactivities.

Radiation chemistry was prosecuted by Milton Burton, Eddie Shapiro, Norman Elliott, and others, not only to see the effect of pile radiation upon water, but also to investigate the stability of solutions that might have importance in nuclear energy: for example, uranium-sulphate solution. George Boyd, Russ Williams, Sol Wexler, and others worked on the Szilard-Chalmers reaction, and found that radiation damage limited the specific activity that could be obtained. Cannon and others developed a calorimetric method for the determination of radiation dose.

Finally, the use of the pile as an analytical tool should be mentioned, inasmuch as we believe that the first use of a reactor for activation analysis was here, with the attendant tremendous increase in the sensitivity of the method as compared with what could be done with weaker neutron sources.

Biology

In its early years, the graphite reactor was the site of an extensive biological program. The most conspicuous part was the irradiation of mice in animal tunnels near the top of the pile. The work sought to differentiate between the effects of fast-neutron, slow-neutron, and gamma radiation using both continuous and intermittent exposures. It even included an experiment on the biological effect of fission particles from colloidal uranium introduced into the animals. The names of Raymond Zirkle, Paul Henshaw, Howard Curtis, and George Stapleton come to mind in association with these activities. The additivity of different kinds of radiation was investigated; LD-50 curves were established, and health-physics tolerance levels were worked out.

In experiments more remote from the pile, ^{32}P plaques were used to observe the effects of external beta radiation upon mice, fern spores, and other biological subjects. This was the first time that such experiments were possible. John Raper joined with others already mentioned, and many observations were made on the pathological effects of this kind of radiation. The induction of tumors and the relative sensitivity of different organs in mice were studied. All kinds of requests came in from other laboratories for pile irradiation of biological material. It was in the graphite reactor that Alan Conger did his work on tradescantia, which showed that the boron content was a major factor in determining the biological effects of slow neutrons. A bismuth box in the thermal column on top of the reactor was used by Stapleton and by Shepard in tangling with dosimetry problems.

Conclusion and in memoriam

That the graphite reactor has been retired after twenty years of faithful service may bring sentimental regrets to those who saw its birth. But this is the inevitable fate of all scientific equipment in an age of rapidly advancing technology. Many who were responsible for bringing the graphite reactor into being have also been responsible for the magnificent technical advances that have made it obsolete. We must therefore look upon the retirement of the graphite reactor not with regret, but with a sense of pride at the many technical achievements over the past twenty years that have made Dr. Doan's final pressure upon the scram button a natural and satisfying event.