

RADIATION EFFECTS

By Frederick Seitz

A research professor in the physics of solids at the University of Illinois at Urbana, the author is chairman of the American Physical Society's Division of Solid State Physics. Before joining the Illinois physics staff, Professor Seitz served for seven years as head of the Carnegie Institute of Technology department of physics.



The following article is based on an introductory presentation during a session of invited papers of the Division of Solid State Physics at the meeting of the American Physical Society at Columbus, Ohio last March.

AS FAR AS I AM AWARE, this is the first invited session of the American Physical Society devoted to the topic of radiation effects in solids—a field that has been an active one for nearly a decade. I believe it is safe to say that it will not be the last such session. The long delay in this event arises, of course, from the fact that the field received its greatest stimulus from the wartime research in the field of atomic energy so that many aspects of it were classified at the start and will remain classified indefinitely in the future.

It will be recalled that by 1942 the group working at the Metallurgical Laboratories of the Manhattan District in Chicago, being reasonably certain that a chain reaction could be initiated using natural uranium and a suitable moderator, began to consider the form which large-scale reactors for the production of plutonium would take. It has always seemed to me that the investigation which ensued at that time on the design of the reactors which would eventually be established at Hanford represents one of the most magnificent technological extrapolations ever carried out by mankind. Even after the first successful chain reaction was staged on December 2, 1942, the group had only pinpoint

amounts of plutonium and the basic knowledge of nuclear physics on which to undertake the design and construction of operating units which would be sufficiently large to produce plutonium in practical quantities. It was necessary to do thinking and planning which represented an extrapolation of the order of a million from the work at Chicago to the pilot plant at Oak Ridge and on through to the production reactors.

During the critical period in 1942 and 1943 when plans were being frozen, the group working in Chicago left no stone unturned in trying to estimate possible difficulties that would arise once the large reactors were set in operation. A host of intricate and novel problems arose and among these were those relating to radiation damage. Both Spedding and Teller pointed out in the middle of 1942 that the presence of high radiation flux might cause changes in the mechanical properties of reactor materials; however, the topic was first given strong coherent recognition by Wigner.

WIGNER RECOGNIZED in the latter part of 1942 that a large fraction of the solid parts of the reactor, most prominently the moderator and the

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fuel elements, would be subject to very heavy bombardment by energetic massive particles such as neutrons and fission fragments. He attempted to decide if this bombardment would give rise to serious technological effects.

Up to this time the problem of radiation effects had been of principal interest to biologists who were concerned with rather subtle changes in life processes and to radiation chemists who were concerned with changes in rate processes, in the radiation-induced reactions of aqueous solutions, or in the coloration of minerals. The nearest comparable studies that had been undertaken previously were the investigations of Lind¹ and his co-workers and of Przibram² on the discoloration of salts and minerals by bombardment with radiations from natural radioactive substances; however, the observed effects were produced by radiation fluxes many powers of ten smaller than those to be expected at Hanford.

In the midst of examination of a host of other perplexing problems, Wigner carried out rudimentary investigations of the fraction of atoms which would be displaced from their normal positions in the graphite moderator in a reasonable period of time and came to the conclusion that the effects could not be ignored. This investigation of what his colleagues eventually termed "the Wigner disease" was followed with similar calculations by Franck, Barton, Szilard, Teller, Metropolis, Morrison, and Friedman which extended the work and confirmed Wigner's views. At the same time James Franck and Milton Burton³ established a group that would investigate the effect with the aid of cyclotron radiations in the period before large-scale reactors were actually available. This group, which contained such outstanding chemists as T. Neubert, A. Novick, B. Leaf, E. Royal, M. Bowman, R. Penneman, and A. O. Allen, not only carried out much work of unusual interest but established standards and techniques which have endured, thereby guaranteeing that the field would be placed upon a firm scientific foundation.

At the request of Franck and with the aid of E. U. Condon, a series of measurements were made at the Westinghouse Research Laboratories by Siegel and Hunter which were among the first to demonstrate that substantial changes in the properties of solids could be produced by nuclear radiations.

W. H. Zachariasen rendered a uniquely valuable service to the program by making the first identification of physical changes with the use of X-ray diffraction. During the second quarter of 1943, a large fraction of the theoretical physicists at Chicago were transferred to Los Alamos and the author was among those recruited to the Chicago laboratories in order to continue the study of Hanford problems. R. J. Maurer also joined the staff at Chicago, and a small group which

had the objective of exploring some long-range aspects of radiation damage in solids was established at the Carnegie Institute of Technology with the cooperation of O. Stern, I. Estermann, and J. S. Koehler. Once the Hanford site became habitable, a group to be concerned with possible practical problems that might arise, was organized at Hanford by the du Pont Company under the guidance of J. A. Wheeler. The problem of radiation damage was part of its field of study. A branch of the radiation group working with Burton was also set up at Oak Ridge once the experimental reactor there began to operate.

Much of the research carried out in the field of radiation damage at Chicago between the beginning of 1943 and the end of the war remains classified because it is concerned with the possible behavior of large-scale operating reactors. It can be said, however, that this study represented, on a small scale, an example of that notable type of extrapolation which was being carried out on a much larger scale on the entire project. As in more important cases, one had a few basic experimental facts obtained at low flux levels and a theoretical structure to serve as a guide for estimating what would transpire at higher fluxes. In the present case, the most important experimental facts were obtained by the chemists and physicists working on the project who examined the very modest changes induced in the physical properties of materials when bombarded with neutrons obtained either from cyclotrons, or from the relatively low flux reactor at Oak Ridge, once it had been set in operation. The guiding theory was two-fold in nature: first was the highly developed theory of the manner in which energetic radiations are dissipated in matter, founded by Bohr early in the history of nuclear physics and extended by many brilliant theoretical physicists; second was the theory of crystal imperfections, still in a rudimentary stage, to be used in connection with the more exact theory to estimate the changes one might expect in solids on an atomic scale as a consequence of the presence of the energetic radiations. From these ingredients, with an ample seasoning of intuition and hope, the group at Chicago attempted to guess what would happen to the solid constituents on a microscopic scale after the large reactors had been placed in operation. Among the conventional fields of science, perhaps only those who are engaged in astrophysics undertake similar extrapolations in their everyday work.

It is a tribute to the group that most of the extrapolations made were correct in a semi-quantitative sense and hence provided a valuable guide to impending operations. Only one or two important effects were judged inadequately, and these are obscure even at present. The speaker believes that these successes, like successes of a similar kind which were achieved during the war, rest upon the fact that a number of individuals having rather highly diverse views concentrated on the problem under circumstances which made it possible for each to contribute his share with a recognized spirit of equality. There was no attempt by any one individual,

or small group of individuals, with a specialized viewpoint to dominate the forecasts. I believe that there is much to be remembered from this history at the present time when our civilization is passing through another great period of crisis and the ability of different segments of society to cooperate for the common good may prove to be the decisive factor.

THE GROUP AT CHICAGO found that the primary effect of neutrons on materials is to produce knocked-on atoms as a result of elastic or inelastic collisions. The greatest changes produced by neutrons occur subsequently as a result of the effects induced by the charged recoil atoms. Such recoil atoms, which behave qualitatively much like natural alpha-particles or fission fragments, have three prominent consequences:

(1) They may excite the electrons in the crystal, producing excitation and ionization. The majority of the energy of the moving particle is lost in this way.

(2) They may transmit energy to the atoms which lie near their track without actually displacing these atoms from their normal position. This action is much the same as if the atoms along the track were raised instantaneously to a very high temperature, of the order of 10^4 ° C. Such "thermal spikes", as they have come to be known, will stimulate any of the effects which high temperatures would. For example, they may promote diffusion in crystals or they may produce disordering. The high temperature is exceedingly transient, lasting only for a time of the order of 10^{-11} seconds, so that the heated wake of the particle is rapidly quenched to ambient temperature. In metals, the excited electrons and the atoms of the lattice come into thermal equilibrium very slowly, so that the degree of heating associated with encounters that set the lattice into vibration directly could not be achieved by exciting the electrons alone. One of the most notable experiments displaying the influence of such thermal spikes is that carried out by Siegel⁶ on the disordering of ordered Cu₃Au alloy by neutron bombardment.

(3) Finally, the knocked-on atom may make sufficiently close encounters with other stationary atoms to displace them from their normal positions, thereby producing particles which can behave much as can the atom struck by the neutron, although their energy is usually much smaller.

The crystal through which the neutron has passed will contain interstitial atoms and vacancies as primary products and will also contain, along the tracks of knocked-on atoms, regions where there has been a high degree of ionization and regions where the lattice has been heated to a very high temperature and rapidly quenched. The ionization may produce observable effects in insulators where electrons and holes may not recombine quickly. The vacancies, interstitial atoms, and heated zones can produce interesting changes in the properties of any of the solids including metals.

It is evident that a material which has been disordered by bombardment with neutrons or other radiations is in a higher energy state than one which has

not been irradiated. Hence it may give off heat when raised to a temperature where the disordering effects anneal. Proper calorimetric studies provide a method of determining the amount of radiation damage.

Fission fragments which behave like knocked-on atoms having an exceedingly high energy, in the vicinity of 80 Mev, will of course produce in a striking way all the effects that neutrons can produce.

Among the effects of radiation should be included those chemical changes which result from transmutation. Such effects are usually masked by changes resulting from ionization, displacement, and heating in the early stages of radiation but may become prominent when a substantial fraction of the material has been transmuted.

The most prominent effects of gamma-rays and fast electrons are those related to excitation and ionization since both primary electrons and those produced by gamma-rays normally have a low momentum and will transfer their energy to the electronic system with overwhelming preference. Mills⁶ has pointed out, however, that the momentum of electrons having energy in the neighborhood of 1 Mev is sufficient to produce some displacement in the lattice. Under proper circumstances this effect may be singled out and studied to great advantage, as has been done by Lark-Horovitz and his coworkers.

IN THE POST-WAR PERIOD there was at first a diminution of effort in the field of radiation damage because many of the principal investigators returned to their pre-war occupations. However, the activity eventually grew again under the stimulation provided by the Atomic Energy Commission. Some of the principal investigators at Chicago, such as Neubert, remained there for a period of time under the leadership of O. C. Simpson, who extended the group by the addition of such individuals as Hennig, Primak, Pringsheim, Delbecq, and Yuster. Similarly, A. O. Allen organized a group of chemists at Oak Ridge. The work at that laboratory was eventually augmented by the temporary or permanent addition of physicists and metallurgists, such as S. Siegel, D. S. Billington, L. P. Hunter, W. E. Johnson, T. Blewitt, W. E. Taylor, O. Sisman, and J. H. Crawford. Hanford was taken over by the General Electric Company which maintained the laboratory established there during the war and extended the work in Schenectady under the stimulating leadership of such individuals as H. Brooks, J. D. Ozerhoff, J. P. Howe, J. R. Low, and more recently, W. W. Tyler and L. Coffin.

In addition, new groups were set up either at universities or at new branches of the Atomic Energy Commission. For example, an Atomic Energy Laboratory was organized at North American Aviation under the direction of C. Starr. There outstanding nuclear physicists such as H. Yockey, W. E. Parkins, and M. M. Mills undertook to use the radiations produced by accelerators in much more refined ways than the wartime groups had attempted. This group has been

greatly stimulated by the addition of individuals such as F. W. Brown, S. Siegel, D. Bowen, and G. J. Dienes.

At Purdue University, Lark-Horovitz⁴ and his associates, including H. M. James, H. Y. Fan, V. A. Johnson, M. Becker, R. E. Davis, and J. Forster, examined the influence of cyclotron radiations on the semiconducting properties of silicon and germanium. This work was supplemented by studies involving reactor radiations in close cooperation with the group at Oak Ridge, most particularly J. C. Cleland, J. H. Crawford, and J. C. Pigg. Investigators at Oak Ridge had independently initiated work on the influence of neutron bombardment on semi-conductors.

Similarly, Brattain and Pearson⁵ of the Bell Laboratories undertook an examination of the influence of alpha-particle bombardment on the properties of germanium.

We may expect a highly productive period from Brookhaven National Laboratory in the near future with the addition of a number of prominent scientists interested in radiation effects such as McReynolds and Dienes.

At the Massachusetts Institute of Technology, Warren and Averbach have been extending the study of the influence of radiation-induced changes on X-ray diffraction, started by Zachariasen, whereas Kaufman and his associates have been concerned with technological aspects of the topic.

Finally, mention should be given to the fact that the subject of radiation effects in solids has been of great interest to investigators in the Canadian and British atomic energy laboratories.

The Atomic Energy Commission in 1949 appointed a committee, of which J. C. Slater was chairman, to undertake a summer study of the field of radiation effects in order to provide the Commission with advice on the way in which it might coordinate and extend its efforts in the field as a whole. The work of this committee has been reviewed by Slater in a recent issue of the *Journal of Applied Physics*.⁶

IF ASKED TO EXPRESS my own views of the scientific future of the field of radiation effects, I believe it would be safe to say that, with suitable control, such studies will provide an exceedingly valuable supplement to those which use a number of other methods of altering the properties of solids, such as raising or lowering the temperature, the application of high pressures or cold work, and the addition of foreign atoms. Moreover, the analysis of radiation effects will yield the richest harvest when employed in conjunction with those other means of controlling or altering solids.

It becomes possible with the use of suitable radiation to introduce determined amounts of vacancies and interstitial atoms in both metals and insulators. If the temperature is maintained sufficiently low during the process, the vacancies and interstitial atoms may be retained in a metastable condition. Thus some of the effects of these imperfections may be studied both

when they are in this metastable state and during migration as the system is subsequently warmed. In this sense the use of radiation may make it possible to alter the density of vacancies in all solids in a manner similar to that in which the density of positive-ion vacancies may be increased in the alkali-halides by the addition of divalent metallic impurities.

Radiation may be used on insulators or semi-conductors to produce new color centers, donor or acceptor levels, and trapping centers without the requirement that the specimen be subject to the more standard and drastic methods of altering the density of levels between the filled and empty bands, such as heating and quenching or remelting.

Minute or even major changes in the chemical composition of suitably chosen solids can be produced by transmutation. This technique makes it possible to study continuously the changes in properties of a single specimen as its chemical composition is altered.

Similarly, appropriately selected radiations may be employed to produce disordering in ordered alloys or to inject free electrons or holes into predetermined regions of an insulator or semi-conductor.

The study of the plastic properties of ductile metals or salts which have been given controlled amounts of irradiation may have very important consequences on the development of our understanding of plasticity. As far as we are aware today, irradiation should not have an appreciable effect on the dislocation pattern of a specimen, at least if carried out at suitably low temperatures where diffusion ceases. On the other hand the products of irradiation may have a strong influence upon the mobility of dislocations which already exist in the crystal. Hence the study of the plastic properties of irradiated materials may well furnish entirely new information on the interaction between dislocations and other types of imperfections and thereby shed new light on the problems of plasticity, as well as on other aspects of solid systems which hinge upon such interactions.

Above all, we should not expect miracles to ensue from relatively haphazard irradiation studies, any more than we should expect miracles from haphazard experiments involving X-ray or neutron diffraction. Crystal-line solids are sufficiently complicated that each method and technique provides the greatest contribution to our understanding of their properties when used to procure that type of information for which it is specifically suited under carefully controlled conditions.

¹ See, for example, Lind, *Chemical Effects of Alpha Particles and Electrons* (Chemical Catalogue Company, 1928).

² K. Przibram, *Zeits. f. Physik* **68**, 403 (1931).

³ M. Burton, *J. Phys. Chem.* **51**, 611 (1947).

⁴ A brief account of the theory is given in the paper by the author, *Discussions of the Faraday Society*, p. 271 (1949). See also K. Lark-Horovitz, *Semiconducting Materials* (Academic Press, Inc., New York, 1951), p. 47; D. S. Billington and S. Siegel, *Metals Progress* **58**, 848 (1950).

⁵ S. Siegel, *Phys. Rev.* **75**, 1823 (1949).

⁶ See the account of M. M. Mills' theoretical work in the review article by K. Lark-Horovitz, reference 4.

⁷ W. H. Brattain and G. L. Pearson, *Phys. Rev.* **80**, 846 (1950).

⁸ J. C. Slater, *Jour. App. Phys.* **22**, 237 (1951).