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To: D. S. Billington

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From: J. B. Trice

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Intra-Laboratory Correspondence
OAK RIDGE NATIONAL LABORATORY
Solid State Division

To: D. S. Billington

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Subject: Four Experiments (Projects) of Some Interest to Radiation Damage Studies

This is a proposal for a research program the purpose of which is to extend the efforts of the present neutron flux work in the Solid State Division in such a way as to achieve the two most important goals of the project. The first goal is the establishment of a radiation damage monitor which can be used for practically all of the in-pile studies of radiation damage to metals. From the practical standpoint such a monitor would serve as a standardization between reactors and between different locations within the same reactor. Irradiation tests to determine changes at a later date in brittleness and elastic moduli are usually performed to understand how a certain structural material may be successfully chosen for high performance in a specific reactor. This is of obvious importance to general reactor programs. The second goal is ultimately related to the first one and is, in fact, a pre-requisite. It is a determination of the ratio of energy loss by collision to energy loss by ionization for a "knocked on" atom traveling through a community of its fellows. Seitz⁽¹⁾ obtains an expression for this ratio as follows:

$$\frac{\left(\frac{dE}{dx}\right)_{\text{collision}}}{\left(\frac{dE}{dx}\right)_{\text{excitation}}} = \frac{Z^{10/3}}{3n} - \frac{m}{M} \cdot \frac{E_r^2}{E^2} \cdot \frac{\text{Log } \frac{E_0}{E_r}}{76.2}$$

(1) Seitz, F., Trans. Faraday Soc. 5, 271 (1949).

where Z , n , m , M are the usual notations for the atomic number, number of free electrons, mass of the electron, and mass of the nucleons, E_r is the Rydberg energy $2E$ is the product of the mass of the electron and the square of the velocity of the displaced atom, E_0 is the energy of the displaced atom and E^* is given by $32Z^{2/3}(m/M)\text{ev}$. For a value of 2 mev for E_0 this ratio is about 30 for copper.

For an elastic collision between a neutron and a heavy atom, the energy transmitted to the heavy atom is given by

$$\Delta E = E_0 (1 - \cos \theta) \frac{2 A}{(A + 1)^2}$$

where A is the atomic number of the target element. For copper at a neutron bombarding energy of 1 mev one gets a maximum energy loss of the neutron of 62 kev and an average energy loss of half this amount or 31 kev. For 14 mev neutrons the maximum loss possible is 860 kev. At very low energies such as 1 mev and much below, theory⁽¹⁾ predicts that most of the energy imparted to the metal lattice is deposited through an elastic collision process which is primarily responsible for distributing the energy of the primary "knock on". For energies of a few mev, however, not all of the energy from the primary "knock on" is distributed through billiard ball collisions.

Some of this is dissipated through transfer to the valence electrons which do not necessarily produce displaced atoms. It is a knowledge of this ratio which we are after since it would inform us as to the necessity for ascribing a certain fraction of energy absorbed during a neutron bombardment to this mode of dissipation. If one requires a radiation damage monitor which measures the neutron bombardment field by means of changes in its physical properties as a result of lattice displacements, then, to trust such a monitor in a different spectral environment it is necessary to know that the monitor sensitivity to neutron energy change parallels the test material's sensitivity.

There is also a considerable interest in this ratio since it is theoretically relevant to a better understanding of radiation damage and displacement theory.

The first suggested experiment is the outgrowth of several discussions with D. O. Thompson, R. H. Kernohan and D. K. Holmes. Thompson and Holmes are presently engaged in an experiment which measures both radiation induced changes in Young's modulus by measurement of the change in resonant frequency and changes in internal friction through measurement of the change in the width of the resonant peak. The apparatus excites a single copper crystal to its normal vibrational modes in situ in the x-pile⁽²⁾. The precision of measurement is very high and the sensitivity is such that highly reproducible changes can be induced by neutron flux irradiations of the order of 10^{11} nvt. The method is therefore somewhat unique, since most other radiation damage measurements in metals either require irradiations of 10^{17} to 10^{20} nvt or else yield non-reproducible results with present experimental equipment and techniques. An attractive experiment to perform with the Young's modulus apparatus is a measurement of change in the modulus as a function of nvt with neutron energy being the varied parameter. Measurements would be made at 0.1, 1, 2, 3, 4 etc. mev and collected data would appear in the form noted in Fig. 1.

Unless the changes in Young's modulus are either rate sensitive or sensitive to the size of displacement spikes (which is another way of saying sensitive to the energy of the primary "knock ons"), a single value of the modulus should correspond to a certain fixed number of defects in the lattice regardless of the particular group of neutrons which caused the damage. A subtraction of the energy values at points A and B, for example, would yield the amount of ionization energy imparted to the sample by neutrons of energy 2.0 mev over those at 0.1 mev.

(2) D. O. Thompson, D. K. Holmes, T. H. Blewitt, "Neutron Irradiation Effects Upon Young's Modulus and Internal Friction Upon Copper", J.A.P., V. 26, #9, 1188 (Sept. 1955).

Fig. 1

Monochromatic Neutrons are obviously not available at reasonably low energies for such purposes from a reactor. However, two other devices are susceptible to exploitation, namely, charged particle reactions with the Van de Graaff and artificial neutron sources. The three most popular sources of neutrons from Van de Graaff machines are the $T + p \rightarrow He^3 + n$ reaction, the $D + D \rightarrow n + He^3$ reaction and the $Li^7(p,n)Be^7$ reaction. The $Li^7(p,n)Be^7$ reaction is a prolific source of monochromatic neutrons and would probably be the logical choice for the presently described work. Energy resolution of the neutrons from this reaction is very high. Since Thompson and Holmes are considering this experiment already, there appears to be no need to dwell on the mechanics of it in this memo. Reference however is made to the comprehensive review article describing Van de Graaff

neutron sources and measurement techniques⁽³⁾ by J. L. Fowler, ORNL and J. E. Brolley, Jr., Los Alamos Scientific Laboratory.

Of the artificial neutron sources, two groups are suggested; photoneutrons using pile produced gamma emitters with either deuterium or beryllium and the somewhat more esoteric source of 14 mev neutrons evoked from the $D(T,n)He^4$ reaction generated within a reactor from a mixture of lithium nitrate + D_2O where the thermal neutron fission of lithium supplies the tritons necessary for the high energy reaction.

Monochromatic neutrons with the energy groups shown in the table below are recommended for studies under 1 mev⁽⁴⁾.

PHOTONEUTRON SOURCES

Source	Half Life	Avg. Neutron Energy (kev)	Flux Yield $n_v = 10^{12}$
$Na^{24} + D_2O$	14.8 h	220	$8.9 \cdot 10^6$
$Na^{24} + Be$	14.8 h	830	$7.9 \cdot 10^6$
$Mn^{56} + D_2O$	2.6 h	220	$4.9 \cdot 10^6$
$Mn^{56} + Be$	2.6 h	100(90%); 300(10%)	$8.4 \cdot 10^7$
$Ga^{72} + D_2O$	14.0 h	130	$3.1 \cdot 10^6$
$Ga^{72} + Be$	14.0 h	-	$5.0 \cdot 10^6$
$In^{116} + Be$	54 m	100(40%); 300(60%)	$1.5 \cdot 10^8$
$Sb^{124} + Be$	60 d	30	$1.9 \cdot 10^7$
$La^{140} + D_2O$	40 h	140	$2.3 \cdot 10^6$
$La^{140} + Be$	40 h	620	$1.5 \cdot 10^6$

(3) J. L. Fowler and J. E. Brolley, "Monoenergetic Neutron Techniques in the 10-30 mev Range", to be published in Rev. Mod. Phys.

(4) D. J. Hughes, "Pile Neutron Research," p. 120.

It is imagined that the photoneutron source could be irradiated as a hollow cylinder for an appropriate period of time and then withdrawn into a shielded region with a geometry such that the Young's modulus apparatus could be centered in the middle of the hollow cylinder and thus be irradiated by a uniform and isotropic flux of neutrons. Consideration of several flux monitors for such an experiment reveals that Pu^{240} is probably the best material to use since its cross section is both accurately known and is rather insensitive to changes in energy. It should be contained in a fission chamber to be placed in close proximity to the Young's modulus apparatus. Such a suitable fission probe has already been developed⁽⁵⁾. The energy imparted⁽⁶⁾ to the Young's modulus specimen is given by the expression

$$\bar{E}_a = \int_0^t n_0 \sigma_s(E_n) \frac{nv(E,t)}{(1+A)^2} dt$$

where $\bar{E}_a$ is the energy which has been imparted to the sample at time t .

In the light of future measurements at higher neutron energies (14 mev for example) it is possible that the modulus measurements at several energies below 1 mev might illustrate a non-linear dependence of Young's modulus on energy absorbed not because of ionization losses but because of differences in character between displacement spikes produced from differences in primary "knock-on" energies. The differences in displacement characteristics would of course have to bear a non-linear relation to the modulus before they would be revealed from this experiment⁽⁷⁾.

(5) J. B. Trice, et al, "High Energy Neutron Flux Measurements in the MTR", (to be published).

(6) For low energies (~1 mev) effects due to inelastic events between neutrons and target atoms are not important.

(7) D. K. Holmes suggested this possibility.

However, it is thought that the major difference in the absorbed energy-damage relation would occur between the low energy neutron groups (~ 1 mev) and the 14 mev group. This phase of the experiment would have to be performed in a location of high thermal neutron flux in order to obtain a high yield from the $\text{Li}(n\alpha)\text{T}$ reaction. The method which uses lithium nitrate dissolved in heavy water (D_2O) has already been successfully tested in the x-pile⁽⁸⁾ by T. Novey. He obtained a flux of 10^6 neutrons (14 mev) $\text{cm}^{-2} \text{sec}^{-1}$ in a thermal flux of 10^{12} n $\text{cm}^{-2} \text{sec}^{-1}$. Perhaps an improvement on his method could be made to yield a high 14 mev flux.

The necessary nuclear data for these experiments exists. The scattering cross sections are, of course, well reported up to 3 or 4 mev for copper and others.

At 14 mev, the total cross section of copper has been measured to be 2.96 ± 0.06 barns⁽⁹⁾ and 2.5 ± 0.1 barns⁽¹⁰⁾. Also the (n_2n) ⁽¹¹⁾⁽¹²⁾ cross section has been measured to be 0.51 ⁽¹¹⁾ barns at 14.1 mev. From these measurements, the energy imparted to copper atoms by 14 mev neutrons can be estimated. Additional measurements probably exist but there was not time for an exhaustive literature survey. Some considerations regarding the anisotropy of the scattering cross section of copper especially at 14 mev remains to be studied.

In conclusion it is thought that the type of experiment described could yield a fruitful field for research and would provide the experimenter with many interesting hours of endeavor.

(8) T. Novey, unpublished Oak Ridge Work (1945).

(9) Coon, Graves and Barschal, Phys. Rev. 88, 562 (1952).

(10) L. S. Goodman, Phys. Rev. 88, 686 (1952).

(11) S. G. Forbes, Phys. Rev. 88, 1309 (1952).

(12) E. R. Graves and R. W. Davis, Phys. Rev. 97, 1211 (1955).

The second experiment proposed is a group of two cross section measurements to be performed using the Van de Graaff. The first is the functional dependence of cross section on energy of the $\text{Al}(n,\alpha)\text{Na}$ reaction which is in current vogue as a fast flux monitor. It is supposed to measure neutrons mostly above 8.1 mev, an effective threshold energy for fission neutrons which has been calculated from very naive considerations based on Bethe's equation for penetration of a charged particle through a coulomb barrier⁽¹³⁾. The second measurement is the cross section functional dependence on energy of the $\text{Ni}^{58}(n,p)\text{Co}^{58}$ reaction. This appeared at first, to be the perfect monitor for long term radiation damage experiments since Co^{58} has a 72 day half life. Also, pile measurements fixed its cross section at about 66 millibarns in a region close to fuel⁽¹⁴⁾. Recent measurements in which it was compared with activities induced in other threshold reactions have indicated a marked sensitivity to intermediate neutrons. It has been established that the apparent effect is not due to thermal neutrons since the cadmium ratio is unity. Perhaps a sensitivity to epithermals enhances its value as a radiation damage monitor but it cannot be accepted prior to a good measurement of its cross section.

It is thought that these two measurements would improve the monitoring situation greatly.

The third and fourth suggested experiments will be only briefly mentioned since they would require both extensive planning and considerable deliberation.

It is suggested that, with an eye to future radiation damage work certain strategic locations in the ORR be mapped with photographic plates before the reactor goes to appreciable power. Such a measurement, performed before the gamma

(13) H. A. Bethe, Rev. Mod. Phys. 9 69 (1937), p. 163).

(14) T. Passel, R. H. Lewis, and R. L. Heath, "MTR Technical Branch Quarterly Report", June 20, 1955, 100-16229, p. 11.

level rises to a prohibitive magnitude, could be tied to a flux monitor which could then be used for future irradiations after the reactor has reached full power operation. Perhaps some consideration might be given to effects on neutron spectra caused by the presence of typical experimental materials.

The final project is a water cooled facility for irradiation of materials in a very high flux. The facility imagined has the necessary mechanism for insertion and removal of a test sample from the reactor during operation. However, it differs from others of its ilk since it would position the sample in place so that cooling water would be in intimate contact with the sample at all times. This would prevent annealing of radiation damage from gamma heat and at the same time it would provide a rapid physical access to samples. It is thought that⁽¹⁵⁾ such a facility might possibly employ water cooled fuel plates to enhance the fast neutron flux. The strongest arguments for such a facility arise from consideration of the irradiation studies which have been made with diamond, quartz, silicon carbide and others which have shown lattice changes with long fast flux exposures. From the monitoring viewpoint, some of these materials show much promise for long exposures in high intensity neutron fields, i. e., $\sim 10^{19}$ nvt. However, previous attempts to establish such monitors have been frustrated because of lack of the experimenters control over the conditions of the experiment.

In summary I think that the first two experiments are the most relevant with regard to improving the radiation damage monitoring situation. The last two experiments are intended to provide a better means for both monitoring and accelerating future radiation damage studies.

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(15) J. C. Wilson, private communication.