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**PROTON SIMULATION OF
DISPLACEMENT EFFECTS INDUCED IN
METALS BY 14-MeV NEUTRONS**

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Abstract

In designing a D-T fusion reactor, one must know the effect of a high flux of 14-MeV neutrons on structural materials. Available laboratory sources of 14-MeV neutrons are not intense enough to achieve realistic flux levels in samples. Bombardment with other particles is one way of simulating the expected neutron flux levels. The energy spectrum of atoms recoiling from collisions with bombarding particles can be calculated from elastic-scattering and nonelastic-reaction data for the incident species. This analysis

shows that 16-MeV protons closely simulate the displacement effects caused by 14-MeV neutrons. In niobium the average damage energy per incident 14-MeV neutron is 2.6×10^{-25} MeV-cm²/atom. Electron-excitation losses and threshold effects have been accounted for. The damage energy deposited per incident 16-MeV proton is 6×10^{-25} MeV-cm²/atom. Protons are a useful unexploited tool for investigating the effects on materials of irradiation with high-energy neutrons.

Introduction

When a fusion reactor first achieves a positive power balance from a controlled thermonuclear reaction, the T(d,n)⁴He reaction will almost certainly be the energy source. The neutron from this D-T fusion reaction carries 14.1 MeV—80 percent of the net reaction energy.

Preliminary design concepts indicate that the reactor's first wall will be subjected to a 14-MeV-neutron flux¹ of about 4×10^{14} neutrons/cm²-sec. There will simultaneously be imposed upon this structure a spectrum of less energetic neutrons, particles from both the plasma

and blanket, and a spectrum of photons. A lithium-containing blanket outside the first wall will breed tritium for continued fueling of the reaction, convert the neutron kinetic energy to heat, and serve as a fluid for heat and fuel transport.²⁻⁴ To achieve reasonable efficiency of thermal conversion, the wall-blanket system must be operated at a temperature as high as is consistent with reasonable safety and reactor lifetime.

The high flux of 14-MeV neutrons is of great concern, and the effects of this flux on structural materials must be established. Available laboratory sources provide

fluxes three orders of magnitude lower than that predicted for the first wall. Experience with various fission-generated spectra is of almost no value. The first wall will see more transmutation reactions and a much harder "primary knock-on atom" (PKA) spectrum than are induced by a fission neutron spectrum. Proposals for a laboratory source of 14-MeV neutrons of appropriate strength share complications of unproven technology, high operat-

ing expense, large tritium inventory, marginal flux, and small accessible volume with high flux.^{5,6}

Simulation techniques are, and will remain for at least several years, the only way to investigate the interaction of high flux and fluence of 14-MeV neutrons with structural materials. This study demonstrates the unique suitability of protons for simulating 14-MeV-neutron displacement effects.

Interactions of 14-MeV Neutrons with Metals

WHERE INTERACTIONS OCCUR

There is no appreciable interaction of neutrons with electrons, and we may safely ignore electrons when discussing primary interactions. In metals of interest for structural applications in a D-T reactor, mean free paths are a few centimeters. Primary interactions occur almost uniformly throughout moderately thick materials, and a small fraction of the incident neutrons will interact within the first wall of the reactor. The 14-MeV neutrons do two things to a metal that are potentially damaging: (1) they initiate nuclear reactions that leave new chemical species, and (2) they produce energetic recoiling atoms.

CHEMICAL CONTAMINATION

Several nuclear reactions occur between 14-MeV neutrons and nuclei. Reactions that result in an alpha particle or a proton introduce at the reaction site a moving helium or hydrogen ion and a recoiling atom with an atomic number lower than the original atom. Unstable

reaction products, such as from $(n, 2n)$ reactions, will spontaneously decay, resulting in new elements. The effect of these transmutation products will be varied. Some may be soluble in the parent lattice and cause only mild alloying effects. Others may not be accommodated so easily. The seriousness of hydrogen born in the lattice depends largely upon the particular alloy system. For many metals and alloys the hydrogen will diffuse so rapidly at the temperatures of interest that it will escape to the surface.

Helium formed by transmutation may be a serious problem. The helium atom does not fit very well in the metal lattice and tends to join other helium atoms and vacancies to form a stable aggregate. This aggregate may move under the influence of stress or temperature gradients or other driving mechanisms and coalesce with other aggregates to form bubbles.^{7,8} It is not yet known how significant this problem will be for materials exposed to a high fluence of 14-MeV neutrons. (In addition to helium formed by transmutation, helium ions will also be injected

into the first wall of the fusion reactor from the reacting plasma and from reactions in the lithium blanket.) The amount of helium produced by 14-MeV neutrons will probably exceed the amount produced by the softer fission-neutron spectrum of the fast breeder reactor. Calculations of the rate of helium production in nickel-containing alloys exposed to fission neutrons have been in serious disagreement with measured helium concentrations.^{9,10} A similar calculation for refractory metals in a fusion-reactor environment must be considered only as an approximate lower limit.

DISPLACED ATOMS

A PKA is created by an incident particle whenever a collision with a lattice-atom nucleus imparts sufficient kinetic energy to the atom. When the energy transferred is below that required to remove an atom from its lattice site and place it in an interstitial site, no displacement occurs. The local temperature of the lattice will be raised by such low-energy collisions. Neutron-nucleus collisions may be elastic or nonelastic.

When the neutron scatters elastically, kinetic energy is conserved, and the recoiling nucleus will have an energy that depends on its mass and the neutron scattering angle. For 14-MeV neutrons most PKA's will have sufficient energy to produce further defects before stopping in the lattice. Neutrons of this energy incident on large nuclei have an associated wavelength comparable to the nuclear dimension. The neutron exhibits wave-like behavior and is diffracted in a manner analogous to light diffracted from a slit.

The elastic angular distribution has strong maxima and minima. This results in some PKA energies being more likely than others.

Nonelastic collisions are defined as collisions in which kinetic energy is not conserved. Included in this category are reactions that result in new elements. In a nonelastic collision the neutron is effectively absorbed by the nucleus, and one or more particles may be subsequently emitted. Some of the energy of the incident neutron goes into raising the potential energy (energy level) of the recoiling nucleus. The combined kinetic energy of the nucleus and neutron is less than for elastic scattering. However, an example given later will show that the average PKA energy resulting from nonelastic processes is much higher than the average energy for an elastic PKA. This will, in general, be the case for 14-MeV neutrons on structural metals because of the tendency for elastic scattering to preferentially occur at small angles, i. e., low energy transfer.

The minimum kinetic energy (E_d) that will produce a single displacement may be estimated by the argument used by Seitz.¹¹ The energy of sublimation of an atom or ion is about 5 eV for a typical metal. Since sublimation occurs from a crystal surface, the binding energy of an interior atom will be about 10 eV. Some energy is required to create the lattice strain necessary to accommodate the atom in an interstitial position. If a lattice atom were moved slowly (adiabatically) from a lattice site to an interstitial position an energy somewhat greater than 10 eV would be required. However, when struck by a neutron, the nucleus

does not follow the path of least resistance nor does it allow time for neighboring atoms to relax. The process is nonadiabatic. Only an average value can be measured for a real crystal since the energy required depends on the two unique sites involved, the path the atom followed between them, and the velocity along that path. A reasonable estimate is that $E_d \approx 25$ eV.

An approximate value of E_d may be obtained experimentally by bombarding the crystal with monoenergetic electrons. When the electron has just enough kinetic energy to displace an atom, only single Frenkel pairs are produced. The number of these defects produced can be measured. By calculating the energy transfer for electrons to the nucleus in question, E_d is deduced. The measured values of E_d for many metals¹² is 22 to 40 eV.

All crystals in thermodynamic equilibrium at a temperature above absolute zero contain vacancies and self-interstitials. These exist because the free energy of the system is made lower by their presence.¹³ Combinations of these elemental defects may exist (i. e., di-vacancy), but at temperatures below one-half the melting point the single vacancy is the only thermodynamically stable point-defect present in significant quantities. The essential feature distinguishing Frenkel defects from those occurring to satisfy thermodynamic considerations is that they come in pairs. Every interstitial produced leaves a vacant lattice site. Vacancies and interstitials are produced in equal numbers.

A moving PKA gives up energy in two ways: (1) it may collide with other lattice atoms, transfer energy, and create addi-

tional displacements, or (2) it may lose energy by exciting electrons.

Energy given up to electrons is removed so rapidly from the "wake" of the moving PKA that lattice atoms are probably not affected by interactions between the PKA and electrons. Energy is dissipated by these interactions that would otherwise cause additional displacements. At PKA energies near E_d , essentially no ionization takes place and little of the energy of the moving atom is lost to electrons. For higher atom velocities (PKA energy), more ionization occurs and a larger fraction of the PKA energy is lost to electron excitation.

The defects produced by a single PKA in the crystal lattice are very concentrated in all but the lightest elements. The cross section for collisions increases rapidly as the PKA slows down, so that near the end of its path the mean free path between collisions approaches the interatomic spacing. Complete derangement of the lattice results.

The final configuration of the atoms in this "displacement spike"¹⁴ is difficult to assess. It will be tens of angstroms across and contain complex clusters of defects as well as Frenkel pairs. Atoms will tend to be ejected from the core leaving a depleted zone¹⁵ surrounded by a region of high interstitial content. Atoms receiving insufficient energy to be displaced may still be heated enough to melt small zones. Microcrystals may be expected to resolidify out of register with the parent lattice.

PECULIARITIES OF A CRYSTAL.

Energy and matter may be transported a considerable distance from a collision

site by several phenomena associated with the crystalline properties of a metal. In a "focused" collision a close-packed row of atoms is smacked on one end in line with the row axis. Under some conditions of energy transfer and effective atom size and spacing, the energy can be transmitted down the row without displacing any atoms. Eventually the energy will be dissipated or an atom will fly off the end of the row.

An atom may also be moved along a close-packed row in a similar concept known as a "dynamic crowdion." If the atom moving along the row axis passes over the equilibrium position it may continue and replace the next in line. This process continues down the row, transporting matter and energy away from the original collision site.

"Channelling" accomplishes a similar effect in a different way. In certain spatial directions a crystal lattice has open channels through which a moving atom can pass. The collisions that the atom experiences are low-angle elastic blows constraining it in the channel.

We see from these arguments that a Frenkel pair need not be a close pair but that the vacancy and associated interstitial may be a considerable distance apart. The effectiveness of these crystalline phenomena is considerably reduced by thermal motion of lattice atoms at higher temperatures and by the ever-present dislocations and other defects in any real crystal.

VACANCY AND INTERSTITIAL MOTION

Only at a temperature of absolute zero will the displacement cascade retain its

original geometry. Surviving point defects are transported through the crystal lattice by diffusion processes. Interstitials are mobile at very moderate temperatures. At temperatures above one-third the melting point, both vacancies and interstitials are migrating freely throughout the crystal lattice. Where they go and how they are accommodated determines their ultimate effect upon the microstructure and mechanical properties of the metal.

Migrating vacancies and interstitials can mutually annihilate by combination or be assimilated at an extended crystal defect such as a crystal dislocation or grain boundary. Interstitials can cluster together to form platelets of extra atoms in the solid. When such platelets contain a few tens of atoms they are more appropriately described as interstitial or dislocation loops. An analogous clustering of vacancies is a dislocation loop of the opposite orientation.

Cawthorn and Fulton¹⁶ reported in 1967 that they observed three-dimensional vacancy clusters in neutron-irradiated stainless steel. Since there is no equivalent interstitial cluster, and matter must be conserved, the existence of these voids within the solid requires that the crystal expand. Such swelling does indeed occur and has since been observed in a variety of metals and alloys¹⁷ including the refractory BCC metals, niobium, vanadium, molybdenum, and tungsten.¹⁸

While some alloy and microstructure details may make a material resistant to swelling, there is no reason to believe that materials will be devised that are immune to swelling. Voids form in the range of temperature from 0.3 to 0.5 of

the melting point, and may contain some gas atoms. They are distinct from bubbles in that the gas present is insufficient for equilibrium between the gas pressure and the surface tension. Helium atoms produced by transmutation were at first thought to serve as sites for heterogeneous nucleation of these defect aggregates.¹⁶ Later experiments substantiate^{19,20} that inert gas atoms

can indeed participate in void nucleation and growth. Remnants of displacement cascades may also serve as effective nucleation sites.

A flood of theories and models have been presented to explain the formation and behavior of voids.^{17,21-23} However, it is relevant to remember that no theorist claims to have predicted the phenomenon of void formation.

What to Simulate

A technique for simulating the effects of 14-MeV neutrons should be capable of providing a rate of atom displacement comparable to that predicted from the D-T reactor flux. It is also desirable to extend displacements per atom to those expected in a reactor lifetime. The simulator should not affect the specimen metallurgy nor interfere with defect kinetics. It must allow use of samples thick enough to be considered bulk. Usual experimental variables of temperature, stress state, and atmosphere should be controllable.

Any reasonable simulation technique must also reproduce the energy spectrum of PKA's. Little is known about the

effect of cascade geometry on kinetics of defects and the fraction of defects that survive. Residual vacancy clusters from energetic PKA's may serve as void nuclei. The PKA spectrum can determine the diffusion distances for defect annihilation or agglomeration. When the PKA energy is only slightly greater than the threshold for displacement, most Frenkel defects are annihilated. Larger fractions of these defects survive when the PKA energy is greater.²⁴ In the absence of an adequate understanding of these effects a useful simulation method must produce a PKA spectrum closely approximating that created by 14-MeV neutrons.

Interaction of 14- to 23-MeV Protons with Metals

The positive charge of a proton causes the proton to interact with electrons in a crystal. Most of the energy deposited by a 14- to 23-MeV proton is delivered to electrons. We need not be concerned with this energy except that it heats the bombarded material.

This discussion of the interaction of protons with nuclei is limited to protons of 14 to 23 MeV incident on medium-weight nuclei. The sample considered must be thin enough so that the energy lost by a proton passing through is small compared to the incident energy.

Protons may initiate reactions leading to new chemical species. The proton, in addition to producing the transmutation elements of lower atomic number that are created by 14-MeV neutrons, may also produce nuclei of higher atomic number.

A proton plowing through the electrons in a lattice will occasionally pass near enough to a nucleus to elastically interact and transfer energy to the lattice atom. The interaction may be dominated by screened-Rutherford, classical-Rutherford, or nuclear forces depending on the energy transferred. The mass of a proton is nearly the same as a neutron. When a proton elastically scatters from a nucleus at some angle, it creates a PKA indistinguishable from one created by a neutron of the same energy scattering at the same angle.

At very low scattering angles the collision is partially "screened" since the proton never gets inside all of the electrons surrounding the nucleus and therefore sees only part of the charge associated with the nucleus. Screening is not an important consideration for 14- to 23-MeV protons. Approach distances large enough to require consideration of screening effects result in energy transfers of less than 1 eV, and will not create primary displacements.²⁵ For a narrow angular range at slightly larger scattering angles, Rutherford scattering predominates. Rutherford scattering is no longer effective when the distance of nearest approach is about equal to the nuclear radius. Nu-

clear forces dominate at small separations, and the proton responds much the same way as a neutron—scattering at preferred angles.

We should expect a "best" simulation for incident proton energies somewhat above 14 MeV for two reasons: (1) Some energy is required to penetrate sufficiently close for nuclear scattering, and (2) the angular region where the scattering mechanism is different for the proton is that where Rutherford scattering predominates. Since the Rutherford differential cross section is inversely proportional to the square of the incident energy, proton scattering at these angles is suppressed by increasing the incident energy.

When the angular distribution of elastic scattering for protons of this energy region is compared to neutron data, close similarities are noted.

Protons also participate in non-elastic collisions that create PKA's. The cross section for proton events of this type is lower than for neutrons of the same incident energy. Other aspects of the preceding discussion of nonelastic neutron interactions are applicable to protons.

The occasional attempts made in the literature at estimating PKA spectra induced by charged particles have assumed that only classical Rutherford scattering occurs.²⁶ Inelastic scattering is lumped at one energy or ignored. This treatment leads to serious errors for protons at energies appropriate for simulation of 14-MeV neutrons.

Ion Bombardment of Samples Thicker than the Ion Range

Many other workers have used charged-particle bombardment to attain high dis-

placement rates in metals. However, such experiments^{26,27} are generally of a

different type than the technique described here. They typically employ the high rate of energy deposition by the bombarding particle near the end of its path to create large numbers of PKA's of low energy. The incident species is consequently stopped in the sample and may chemically interact with the parent material.

Ions of mass quite different from that of a neutron are frequently employed. The resulting PKA spectrum and total displacement rate vary rapidly as a function of the distance from the sample surface, and at no point do they resemble a neutron-induced PKA spectrum.²⁷

Bombardment with heavy ions creates a large number of displacements per atom, but it does not in any other way simulate neutron displacement effects. Advocates of heavy-ion bombardment usually point out that this technique can reproduce the average PKA energy. Duplicating the average PKA energy alone is not a sufficient basis for a simulation technique, since vastly different spectra may have identical average energy. Heavy ions additionally have ranges so short as to preclude testing of bulk samples.

PKA's from Elastic Interactions

When a particle of mass m_1 and kinetic energy E_0 elastically scatters from a nucleus of mass m_2 , the energy transferred to the nucleus is given by

$$E_4 = E_0 \frac{4m_1 m_2}{(m_1 + m_2)^2} \sin^2(\theta/2), \quad (1)$$

where θ is the center-of-mass scattering angle of the incident particle. A PKA is created when this energy exceeds that required to remove an atom from a lattice site and place it in an interstitial site, E_d . ($E_d \approx 35$ eV for many metals.) The maximum possible energy transfer $E_{\max}$ (180-deg scatter) is given by

$$E_{\max} = E_0 \frac{4m_1 m_2}{(m_1 + m_2)^2}. \quad (2)$$

The assumption is frequently made in radiation-damage research that neutron

elastic scattering is isotropic. This assumption is not generally accurate for an incident neutron energy exceeding 1 MeV. At 14 MeV the elastic angular distribution for neutron scattering from refractory metals is strongly anisotropic.

Measured angular distributions and cross sections for 14-MeV elastic neutron scattering are available for most elements. The equivalent information for 14- to 23-MeV protons is measured for some energies and calculable using optical-model analysis.²⁸ Since for elastic events the energy of a PKA is defined by Eq. (1), the portion of the PKA energy spectrum attributable to elastic scattering is defined by the magnitude and angular distribution of elastic scattering for the incident particle. The differential cross section $d\sigma/dE$ for elastically creating a PKA at energy E_4 is directly proportional to the differential cross section $d\sigma/d\Omega$ for an incident particle

elastically scattering at the corresponding angle, where σ is the microscopic cross section and Ω is the solid angle. From Eqs. (1) and (2)

$$dE = (E_{\max}/2) \sin \theta d\theta \quad (3)$$

and

$$d\Omega = 2\pi \sin \theta d\theta, \quad (4)$$

so

$$d\sigma/dE = (d\sigma/d\Omega) (4\pi/E_{\max}). \quad (5)$$

For $d\sigma/d\Omega$ in $\text{cm}^2/\text{steradian}$ and $E_{\max}$ in MeV, $d\sigma/dE$ is the number of primary recoils per atom-MeV for each incident particle.

PKA's from Nonelastic Collisions

Collision events in which kinetic energy is not conserved will be referred to here as nonelastic. For our purposes such a collision can be visualized in two steps. First the incident particle hits the nucleus and sticks. To conserve momentum, the velocity v_g of the nucleus and the incident particle at this point must be

$$v_g = \frac{(2E_0 m_1)^{\frac{1}{2}}}{m_1 + m_2}, \quad (6)$$

which is

$$v_g = \frac{1}{2} v_{\max}, \quad (7)$$

where $v_{\max}$ is the energy corresponding to the maximum possible elastic PKA energy $E_{\max}$. This moving composite then ejects a particle of mass m_1 and kinetic energy E_3 at a center of mass angle θ from the original incident direction. The resulting velocity v_4 of the nucleus may be larger or smaller than v_g depending on θ . The velocity increment δv given to the nucleus by this ejection is

$$\delta v = \frac{m_1}{m_1 + m_2} \left[\frac{2(E_0 - U)}{m_1} \right]^{\frac{1}{2}}, \quad (8)$$

where U is the difference between the energy of the incident particle and the energy of the ejected particle, $(E_0 - E_3)$. Then

$$v_4^2 = (-\delta v \cos \theta + \frac{1}{2} v_{\max})^2 + (\delta v \sin \theta)^2, \quad (9)$$

and the kinetic energy E_4 of the recoiling nucleus from a nonelastic collision is

$$E_4 = \frac{m_1 m_2}{(m_1 + m_2)^2} \{2E_0 - U - 2[E_0(E_0 - U)]^{\frac{1}{2}} \cos \theta\}. \quad (10)$$

Since the time interval between collision and ejection is very small compared to particle travel time for one interatomic distance, the collision and ejection may be considered as occurring simultaneously at one lattice site.

Grimes et al. have shown that for protons²⁹ and neutrons^{30,31} the differential reaction cross section integrated over angle for emission of particles of energy E_3 is accurately represented at incident energies greater than 10 MeV by

$$d\sigma_R/dE_3 = E_3^\alpha \exp(U/T_n) + E_3^\beta U^n, \quad (11)$$

where α and β are nuclear constants reflecting the relative magnitude of equilibrium and pre-equilibrium emission, T_n is the nuclear temperature, and n is 2. This differential cross section is for all nonelastic events that emit a proton or neutron. That is, for neutrons it accounts for (n,n') , (n,p) , and the first neutron from $(n,2n)$ reactions. The equilibrium emission is observed to be isotropic in the center of mass, the pre-equilibrium emission is symmetric about $\theta = 90$ deg.

The fraction F of the reaction cross section attributable to pre-equilibrium emission can be experimentally evaluated.

From Eq. (11),

$$\beta = \frac{F\sigma_R}{\int_0^{E_0} E_3 (E_0 - E_3)^n dE_3}, \quad (12)$$

where σ_R is the integrated reaction cross section. This integrates to

$$\beta = \frac{F\sigma_R (n+1)(n+2)}{E_0^{n+2}}. \quad (13)$$

Similarly for α

$$\alpha = \frac{(1-F)\sigma_R}{T_n^2 \{[\exp(E_0/T_n)] - (E_0/T_n) - 1\}} \quad (14)$$

Equations (11), (13), and (14) define $d\sigma_R/dE_3$ at any value of E_3 for known values of T_n , n , σ_R , and F .

For calculating the PKA spectrum resulting from nonelastic events we may assume the particle emission to be iso-

tropic. While this is not rigorously true for pre-equilibrium emission, the effect on the PKA spectrum of the departure from isotropic emission is negligible. This is true because this emission is symmetric and the effect of the entire pre-equilibrium process on the PKA spectrum is not large. We may also neglect the very small velocity increment given to the nucleus from gamma emission and the second neutron from $(n,2n)$ reactions. Their effect is to introduce a negligible additional broadening in the nonelastically created PKA spectrum. The velocity increment given to a PKA by emission of an alpha particle is larger than that which results from emission of a proton or neutron because of its greater mass. However, the (n,α) reaction represents a small fraction of the nonelastic cross section for materials of interest, so the effect on the PKA spectrum is to create small number of PKA's of energies greater than $E_{\max}$. This effect may also be neglected.

Since the particle emission is assumed to be isotropic it is seen from Eq. (10) that nonelastic collisions in which the kinetic energy loss is U create $d\sigma_R/dE_3$ amount of PKA's, which are equally distributed between energies

E_f and E_b , where

$$E_f = (E_{\max}/4E_0) \{2E_0 - U - 2[E_0(E_0 - U)]^{1/2}\}, \quad (15)$$

and

$$E_b = (E_{\max}/4E_0) \{2E_0 - U + 2[E_0(E_0 - U)]^{1/2}\}. \quad (16)$$

The PKA spectrum created by nonelastic collisions is the sum of these spectra as U goes from $E_{\max}$ to E_0 . The cross sec-

tion for events with $U < E_{\max}$ is small and is included with the elastic cross section.

PKA and Damage Spectra

The total PKA spectrum in a metal resulting from particle bombardment is the sum of the elastic and nonelastic contributions.

Not all of the energy transferred to lattice atoms is available to create displacements. Some PKA energy is given up to nondamaging electron excitation. Elastic collisions at very small angles do not transfer sufficient energy to displace an atom. Kinchin and Pease³² in their early review suggested that the number of displacements N produced by a PKA of energy E_4 is

$$N = \begin{cases} 0 & \text{for } 0 \leq E_4 < E_d \\ i & \text{for } E_d \leq E_4 < 2E_d \\ E_4/2E_d & \text{for } 2E_d \leq E_4 < L_c \\ L_c/2E_d & \text{for } E_4 \geq L_c \end{cases} \quad (17)$$

The energy L_c is defined as the energy of a moving PKA in the lattice above which all energy is lost in electronic excitation and below which all energy is used to create displacements. For metals,³³

$$L_c \approx m_2 \text{ keV}. \quad (18)$$

This energy approximately corresponds to the onset of ionization. Such an abrupt threshold for a change in the energy-loss mechanism does not in reality occur, however this useful fiction has been re-

peated for so long it has joined the folklore of radiation damage.

More recently Linhard³⁴ has derived expressions for the fraction of PKA energy lost to electron excitation as a function of PKA energy. Robinson³⁵ has shown that the Linhard's analysis can be represented by the expression

$$S = k_L g / (1.0 + k_L g), \quad (19)$$

where S is the fraction of the PKA energy lost to electron excitation, g is a functional variable of the ratio E_4/E_L , and E_L and k_L are parameters of Linhard's theory. For an energetic atom moving through identical atoms

$$k_L = 0.133745 Z^{2/3} / m_2^{1/2} \quad (20)$$

and

$$E_L = 86.931 Z^{7/3} \text{ eV}, \quad (21)$$

where Z is the atomic number of the atom whose mass is m_2 . The function g is approximated by³⁵

$$g = 3.4008(F_4/E_L)^{1/6} + 0.40244(E_4/E_L)^{3/4} + (E_4/E_L). \quad (22)$$

This efficiency is plotted in Fig. 1 for a niobium PKA in a niobium lattice. Since the Linhard efficiency accounts only for

Fig. 1. Linhard damage efficiency (fraction of recoiling-atom energy that is available for creating displacements) as a function of initial PKA kinetic energy for a niobium PKA in niobium. (PKA stands for "primary knock-on atom.")

Fig. 2. Damage efficiency at low PKA energy—near the threshold for displacement in niobium.

electron excitation losses, some account must also be made for the displacement threshold. The exact nature of this

transition is unknown and indeed probably varies with temperature, local stress, defect density, and crystallographic direction. When a material is bombarded with electrons of an energy just sufficient to create single defects, a large fraction of the defects are observed to be close pairs that annihilate at temperatures just sufficient to allow interstitial mobility. In this analysis no attempt was made to account for defect annihilation. A sharp displacement threshold and Linhard efficiency at all PKA energies above E_d was assumed. Linhard efficiency and the modified efficiency are plotted in Fig. 2 for this low-energy region.

By taking the number of PKA's at each energy times that energy and times the efficiency for creating displacements, it is possible to create a "damage-energy spectrum," which represents the amount of energy available for creating displacements as a function of PKA energy. This is the heart of the matter. The damage-energy spectrum for the simulating particles is compared to the damage-energy spectrum for the neutrons to evaluate the accuracy of the 14-MeV-neutron simulation technique. Further, the integral of these spectra over the range of PKA energies is the total amount of energy per average incident particle that is available to create displacements. This integral then represents the relative effectiveness of an incident particle for creating displacements.

Results for Niobium

Input required to compute the PKA and damage-energy spectra resulting from 14-MeV neutrons and variable-energy proton bombardment is:

- Mass, charge, and nuclear temperature of the bombarded nucleus
- Neutron differential elastic cross sections
- Neutron nonelastic reaction parameters, F and n
- Neutron nonelastic cross section, σ_R
- Proton incident energy
- Proton differential elastic cross sections
- Proton nonelastic reaction parameters, F and n
- Proton nonelastic cross section, σ_R

I wrote a computer code that, using this input, calculates and displays the:

- Neutron elastic PKA spectrum
- Proton elastic PKA spectrum
- Neutron nonelastic PKA spectrum
- Proton nonelastic PKA spectrum
- Neutron total PKA spectrum
- Proton total PKA spectrum
- Neutron damage-energy spectrum
- Proton damage-energy spectrum
- Damage energy per incident neutron and proton.

The structural material in the first wall of a D-T fusion reactor must be compatible with hot lithium, have suitable high-temperature mechanical properties, and be weldable and nonmagnetic. It should also have low sputtering rates for surface bombardment, low activation

from the radiation environment, and small removal cross section for 14-MeV neutrons. Candidate materials are vanadium, niobium, and molybdenum, and their alloys. Niobium has been the most studied and seems the most promising as the major constituent for the first wall material.^{4,8} I have collected published data and applied this analysis to calculate the accuracy of using protons to simulate 14-MeV neutron displacement effects in niobium.

Kammerdiener measured differential elastic cross sections³⁶ of 14-MeV neutrons on niobium at the Livermore ICT accelerator. These measurements extended from $\theta = 26$ deg to $\theta = 146$ deg. I calculated Wick's limit³⁷ for scattering near 0 deg, and treated the cross section above 146 deg as constant at the same level. These cross sections are plotted in Fig. 3. They are not markedly different

Fig. 3. Elastic scattering on niobium. Differential elastic cross sections for 14.1-MeV neutrons and 16.4-MeV protons.

from the synthetic ENDF B³⁸ function form used by Robinson.³⁵

Nonelastic reaction parameters for 14-MeV neutrons on niobium^{29-31,30} are: $F = 0.15$, $n = 2$, and $T_n = 0.68$ MeV. The value of σ_R is 2 b.⁴⁰

I wanted the capability to calculate proton-induced PKA spectra for arbitrary incident proton energy. Since experimental data are not available for all interesting energies, I used an optical-model scattering code to generate the required proton input. The original authors of the optical-model code were Fernbach and Bjorklund at LLNL about 1957. Input parameters as suggested by Becchetti for best fits to proton data were used.⁴¹ Comparisons to published proton-scattering data were made for 16.4- and 22.4-MeV protons on niobium;⁴² and for 14-MeV protons on molybdenum.⁴³

Figures 4 and 5 present calculated and experimental values of the ratio of elastic proton scattering to hypothetical classical Rutherford scattering for niobium. The 16.4-MeV data are the same as presented in Fig. 3. I concluded that the optical-model calculation was sufficiently precise to serve as input to the PKA calculations. The same optical-model code also provides the proton nonelastic cross section σ_R . Figures 4 and 5 also illustrate the error in assuming that protons scatter only by Rutherford interactions. Data equivalent to these were published for niobium in 1954.⁴⁴

Nonelastic reaction parameters for protons of this energy on niobium²⁹⁻³¹ are: $F = 0.10$, $n = 2$.

The preference for low-angle elastic scattering for both protons and neutrons creates relatively larger numbers of low-

Fig. 4. Scattering of 16.4-MeV protons on niobium. Comparison of optical-model calculation of elastic cross section with published measurements⁴² at 16.4-MeV incident energy.

Fig. 5. Scattering of 22.4-MeV protons on niobium. Comparison of optical-model calculation of elastic cross section with published measurements⁴² at 22.4-MeV incident energy.

energy PKA's. The average PKA energy for both spectra (Fig. 6) is below 100 keV. The maximum energy PKA from a neutron

Fig. 6. Elastic PKA spectra in niobium. Energy spectra of recoiling niobium atoms from elastic collisions with 14.1-MeV neutrons and 16.4-MeV protons.

collision is 590 keV. Since the proton energy is somewhat higher than 14 MeV, PKA's up to 690 keV are born from proton collisions.

From Eq. (11) the most likely non-elastic event is for an ejected particle with near zero energy. This results in a PKA of energy equal to one-fourth the maximum. The nonelastic cross section for 16.4-MeV protons is 1.1 b. This is reflected in Fig. 7 by fewer recoils per incident particle when integrated over PKA energy.

The total PKA spectra (Fig. 8) is the sum of the elastic and nonelastic components. The local maximum at one-fourth the maximum PKA energy created by nonelastic events dominates the spectra near that energy. At energies very far from this region, elastic events determine the total spectrum.

The threshold displacement energy E_d for niobium is 36 eV.⁴⁵ The damage-energy spectra presented in Fig. 9 are generated by applying the efficiency of

Fig. 2 to the total PKA spectra. The integral of the neutron-induced damage-energy spectrum is $2.5 \cdot 10^{-25}$ MeV-cm² atom-(incident neutron).

The total damage energy for an incident proton is sensitive to the value of E_d chosen. Quantitative results must be considered in the light of the uncertainty about survival of defects created by low-energy PKA's. Protons of these energies deliver about twice as much energy to the lattice as 14.1-MeV neutrons. The amount of damage energy decreases slightly with increasing incident energy because of the suppression of small-angle Rutherford scattering. The damage-energy spectra for various-energy protons in Fig. 10 reflect this effect. The value of E_{max} and the local maximum at $1/4 E_{max}$ shift to higher energies with increasing incident energies. The integrals of these damage-energy spectra from various lower limits to E_{max} are presented in Table 1.

Fig. 7. Nonelastic PKA spectra in niobium. Energy spectra of recoiling niobium atoms from nonelastic collisions with 14.1-MeV neutrons and 16.4-MeV protons.

Fig. 8. Total PKA spectra in niobium. Energy spectra of recoiling niobium atoms from collisions with 14.1-MeV neutrons and 16.4-MeV protons.

Fig. 9. Damage-energy spectra in niobium. Damage-energy density in niobium versus PKA energy for 14.1-MeV neutrons and 16.4-MeV protons.

Fig. 10. Damage-energy spectra in niobium. Damage-energy density in niobium versus PKA energy for various incident proton energies.

Table 1. The integral from $E_{\max}$ to various lower limits of the damage-energy spectra plotted in Fig. 10.

Lower limit of integration (keV)	Damage energies (barn-keV)				
	Neutrons	Protons			
	14.1 MeV	14.1 MeV	16.4 MeV	19.0 MeV	22.4 MeV
593.6	0	0	2	6	14
500.0	6	4	8	13	23
100.0	222	141	165	190	223
10.0	254	259	260	266	282
1.0	258	414	393	381	379
0.5	258	463	435	417	410
0.036	258	655	600	560	531

Discussion and Limitations

Protons at 16 MeV accurately simulate the 14-MeV-neutron damage-energy spectra over most of the PKA energy range. The only significant difference occurs at energies between E_d and about $30E_d$. At temperatures of interest for a D-T reactor, most of the defects created by PKA's in this energy region will not contribute to the radiation damage process. Protons are not suitable for simulation of neutrons with lower energy, where proton elastic scattering is almost completely Rutherford scattering. The simulation also becomes less accurate at lower temperatures since low-energy PKA's then affect the number density of surviving defects.

A program to investigate the fundamental basis of damage caused by neutrons and to experimentally evaluate the accuracy of proton simulation is possible using existing equipment and techniques.⁴⁶

The damage energy deposited per unit volume by a given flux can be calculated from the integrals presented in Table 1. Since each integral is the product of microscopic cross section and energy, it must be multiplied by atom density and flux to obtain the damage energy per unit volume. If flux is in particles/cm² and atom density in atoms/cm³ then the integral in Table 1 must be converted to cm²-keV to get damage-energy deposition in keV/cm³.

Other Aspects of Proton Bombardment

The prospect of using protons as neutron simulators is attractive because protons have several experimental advantages over neutrons.

The Livermore ICT accelerator and target system is the most intense source of 14-MeV neutrons available. It provides a flux equivalent to 1×10^{12} neutron/cm²-sec

through an accessible volume large enough to expose small-bulk material samples. The existing LLL Cyclograaff (a combination of a cyclotron and a tandem Van de Graaff accelerator) can deliver $10 \mu\text{A}$ of protons at 16 to 27 MeV. By focusing the beam to an area of 0.2 cm^2 , a sample can be exposed to a flux of 3×10^{14} protons/ cm^2 -sec. A flux of 10 times this magnitude is attainable by taking the beam directly from the cyclotron at 15 MeV.

Proton-irradiation experiments are easily adapted to mechanical testing under bombardment. The beam can be transported to the experiment and delivered to the sample without exposing the test equipment and instrumentation to direct beam flux. Constant-strain-rate, constant-stress, stress-relaxation, and fatigue tests could be accommodated with controlled conditions of atmosphere, temperature, flux, and fluence. Experiments designed to study high-fluence effects such as voids and other complex microstructure would require long-term exposure. A fluence of 1×10^{23} protons/ cm^2 could be delivered in 1 year of nonstop operation.

In order to preserve the similarity in the primary knock-on spectrum, the sample thickness for proton bombardment must be chosen so that the energy lost by the protons in passing through the sample

is small compared to the incident energy. At high current density a more restrictive requirement on sample thickness is the removal of the heat deposited by the passage of the proton beam. At a beam current density of $500 \mu\text{A}/\text{cm}^2$ on niobium a sample thickness of 1.5 to $15 \mu\text{m}$ could be exposed. The protons would lose 0.02 to 0.2 MeV in passing through the sample.⁴⁷ Several samples could be bombarded in sequence in the same beam with successive loss of energy and beam intensity.

Most protons in this type of exposure would pass through a sample without creating a PKA. Of those which elastically scatter only a few would come to rest in the sample. For a flux of 3×10^{14} protons/ cm^2 -sec on a $50\text{-}\mu\text{m}$ foil of niobium, about 10^{-8} atom fraction per day of hydrogen would be elastically scattered into the solid. Nonelastic nuclear collisions affect 10^{-4} of the incident protons.⁴⁷ Most of these events will not result in a proton.⁴⁸ The average range of a proton emitted from a reaction will be several foil thicknesses. It would take several years to implant 1 ppm (by weight) of hydrogen at these rates. Furthermore, the diffusion of hydrogen in niobium is so rapid⁴⁹ that the steady-state concentration under bombardment would be negligible for much higher implantation rates.

Summary

In collisions of 14-MeV neutrons with medium-weight nuclei, nonelastic events create PKA's of higher average energy than elastic interactions and produce more defects than elastic interactions. In niobium the average damage energy per inci-

dent 14-MeV neutron is $2.5 \times 10^{-25} \text{ MeV-cm}^2/\text{atom}$. The average damage energy per collision is

$$\frac{2.6 \times 10^{-25} \text{ MeV-cm}^2/\text{atom}}{4.0 \times 10^{-24} \text{ cm}^2/\text{atom}} = 65 \text{ keV}.$$

The average niobium PKA created by a 14-MeV neutron carries 65 keV, which will eventually be utilized to create defects. Protons with 16-MeV energy closely simulate displacement effects caused by 14-MeV neutrons. They are available at much higher fluxes than

14-MeV neutrons and offer other experimental advantages.

This study demonstrates that protons are a useful tool for investigating some aspects of radiation damage anticipated in a D-T fusion reactor.

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References

1. J. D. Lee, Estimate of Radiation Fluxes Expected to Hit First Wall of DT and D^3He Mirror Reactors, Lawrence Livermore Laboratory, Rept. UCID-15944 (1971).
2. D. J. Rose, On the Feasibility of Power by Nuclear Fusion, Oak Ridge National Laboratory, Tennessee, Rept. ORNL-TM-2204 (1968).
3. J. D. Lee, "Tritium Breeding and Energy Generation in Liquid Lithium Blankets," in Proc. Brit. Nucl. Energy Soc. Conf. Nucl. Fusion Reactors, Abingdon, 1969 (UKAEA Culham Laboratory, Abingdon, England, 1970), pp. 471-482.
4. D. Steiner, Nuclear Appl. & Tech. **9**, 83 (1970).
5. D. Colombant, High Intensity 14 MeV Neutron Source, Ph.D. thesis, Massachusetts Institute of Technology, Cambridge, Mass. (1969).
6. H. Dreicer and D. B. Henderson, Facility for Duplicating 14 MeV Neutron Effects in Fusion Power Reactors, Los Alamos Scientific Laboratory, New Mexico, Rept. LA-4709-MS (1971).
7. D. G. Martin, "An Assessment of Some Radiation Damage Effects in the Containment Vessel of a Thermonuclear Reactor," in Proc. Brit. Nucl. Energy Soc. Conf. Nucl. Fusion Reactors, Abingdon, 1969 (UKAEA Culham Laboratory, Abingdon, England, 1970), pp. 399-409.
8. D. G. Martin, J. Nucl. Mater. **33**, 23 (1969).
9. N. D. Dudgey and S. D. Harkness, Nucl. Appl. & Tech. **9**, 700 (1970).
10. A. A. Bauer and M. Kangilaski, J. Nucl. Mater. **42**, 91 (1970).
11. F. Seitz, Discussions Faraday Soc. **5**, 271 (1949).
12. B. T. Kelly, Irradiation Damage to Solids (Pergamon Press, New York, 1966), p. 2.
13. R. A. Swalin, Thermodynamics of Solids (John Wiley & Sons, Inc., New York, 1962) pp. 214-271.
14. J. A. Brinkman, J. Appl. Phys. **25**, 961 (1954).
15. A. K. Seeger, "On The Theory of Radiation Damage and Radiation Hardening," in Proc. U.N. Intern. Conf. Peaceful Uses of At. Energy, 2nd, Geneva, 1958 (United Nations, Geneva, 1958) vol. 6, p. 250.
16. C. Cawthorn and E. J. Fulien, Nature **216**, 375 (1967).
17. S. D. Harkness and Cui-Yu Li, Metallurgical Transactions **2**, 1457 (1971).
18. J. D. Ekan, G. Hamburg, and A. Mastenbroeck, J. Nucl. Mater. **39**, 194 (1971).
19. E. E. Bloom and J. O. Stiegler, J. Nucl. Mater. **36**, 331 (1970).
20. D. I. R. Norris, J. Nucl. Mater. **40**, 66 (1971).
21. V. F. Sears, J. Nucl. Mater. **39**, 18 (1971).
22. H. Wiendersich, Radiation Effects **12**, 111 (1972).
23. R. W. Powell and K. C. Russell, Radiation Effects **12**, 127 (1972).

24. D. Schmidt, Radiation Damage from Internal Alpha Decay in Thorium, Lawrence Livermore Laboratory, Rept. UCRL-51009 (1971).
25. N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **18**, No. 8 (1948).
26. R. S. Nelson, "Cyclotron Bombardment to Simulate Fast Neutron Damage in Reactor Materials," in Proc. Int. Conf. on the Use of Cyclotrons in Chemistry, Metallurgy, and Biology, Oxford, England, 1969 (Hilife Press, Oxford, 1969).
27. R. S. Nelson, D. J. Mazey, and J. A. Hudson, J. Nucl. Mater. **37**, 1 (1970).
28. F. G. Perey, Phys. Rev. **131**, 745 (1963).
29. S. M. Grimes, J. D. Anderson, B. A. Pohl, J. W. McClure, and C. Wong, Phys. Rev. C **4**, 607 (1971).
30. J. D. Anderson, Lawrence Livermore Laboratory, private communication (1972).
31. S. M. Grimes, Lawrence Livermore Laboratory, private communication (1972).
32. G. H. Kinchin and R. S. Pease, Reports on Progress in Physics **18**, 1 (1955).
33. L. T. Chadderton, Radiation Damage in Crystals (Barnes and Noble, 1965), p.31.
34. J. Linhard, V. Nielsen, M. Scharff, and P. V. Thomsen, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, No. 10 (1963).
35. M. T. Robinson, "The Energy Dependence of Neutron Radiation Damage in Solids," in Proc. Brit. Nucl. Energy Soc. Conf. Nucl. Fusion Reactors, Abingdon, 1969 (UKAEA Culham Laboratory, Abingdon, England, 1970), pp. 364-378.
36. J. L. Kammerdiener, Neutron Spectra Emitted by ^{239}Pu , ^{235}U , Pb, Nb, Ni, Fe, Al, and C Irradiated by 14 MeV Neutrons, Lawrence Livermore Laboratory, Rept. UCRL-51232 (1972).
37. G. C. Wick, Phys. Rev. **75**, 1459 (1949).
38. ENDF/B signifies Evaluated Nuclear Data File. This file is on tape and is maintained by Brookhaven National Laboratory, Upton, New York.
39. V. B. Anufrienko, B. V. Devkin, G. V. Kotelnikova, Yu. S. Kulabukhov, G. N. Lovchikova, O. A. Sal'nikov, L. A. Timokhin, V. R. Trubnikov, N. I. Fetisov, J. Nucl. Phys. (U.S.S.R.) **2**, 826 (1965).
40. M. D. Goldberg, S. F. Mughabghab, S. N. Purohit, B. A. Magurno, and V. M. May, Neutron Cross Sections, Volume IIB, Z = 41 to 60, Brookhaven National Laboratory, Upton, New York, Rept. BNL 325 2nd ed. Supp. No. 2 (1966).
41. F. D. Becchetti, Jr., and G. W. Greenlees, Phys. Rev. **182**, 1190, (1969).
42. C. B. Fulmer, Phys. Rev. **125**, 631 (1962).
43. H. F. Lutz, D. W. Heikkinen, and W. Bartolini, Phys. Rev. C **4**, 934 (1971).
44. B. L. Cohen and R. V. Neidigh, Phys. Rev. **93**, 282 (1954).
45. G. Youngblood, S. Myhra, and J. W. DeFord, Phys. Rev. **148**, 1101 (1969).
46. R. J. Borg, Lawrence Livermore Laboratory, private communication (1972).

47. J. F. Janni, Calculations of Energy Loss, Range, Pathlength, Straggling, Multiple Scattering, and the Probability of Inelastic Nuclear Collisions for 0.1 to 1000 MeV Protons, Air Force Weapons Laboratory, Kirtland AFB New Mexico, Rept. AFWL-TR-65-150 (1966).
48. R. A. James, Phys. Rev. 93, 288 (1954).
49. L. A. Charlot and R. E. Westerman, J. Nucl. Mater. 41, 227 (1971).

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