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UCRL-74675
PREPRINT

Conf-730416-3

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**SIMULATION OF 14-MeV NEUTRON DAMAGE TO
POTENTIAL CTR MATERIALS**

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May 29, 1973

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This paper was prepared for publication in Proceedings of the 2nd International Solid State Conference, American University of Cairo, Cairo, Egypt, April 20-27, 1973.

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SIMULATION OF 14-MeV NEUTRON DAMAGE TO POTENTIAL CTR MATERIALS*

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ABSTRACT

The reaction $D(T, n)^4\text{He}$ occurs either as the primary plasma reaction or as a side reaction in nearly all the controlled thermonuclear reactor (CTR) design schemes. Neutrons produced by this reaction possess an energy of 14.1 MeV and are capable of causing an enormous amount of damage, the extent and character of which is still mostly unknown.

Current CTR design concepts anticipate fluxes of 10^{14} to 10^{15} n/cm²/sec which, except for EBR-II, are quite outside our present technological experience. Hence, from the standpoint of both energy and intensity, CTR neutron sources are expected to create materials problems in an area that is still virtually unexplored. The difficulties in researching these problems are compounded by the obvious lack of an experimental neutron source of comparable strength. This lack forces the materials scientist to employ simulation rather than direct measurement as a means of obtaining the desired information.

In this paper we briefly review the major CTR designs being considered with special reference to the problem of neutron damage.

* Work performed under the auspices of the United States Atomic Energy Commission.

Subsequently, we present a review of the aims and methods of the simulation program in progress at the Lawrence Livermore Laboratory. This program centers about the Rotating Target Apparatus, which is currently the world's most powerful source of 14-MeV neutrons capable of continuous production of approximately 10^{12} n/cm²/sec. This machine is described in detail as are the preliminary results of our attempt to correlate and compare 14-MeV damage to V, Nb, Mo, and Cu with proton and reactor neutron damage.

Most of the world's conventional sources of energy are either fully developed, approaching a foreseeable state of exhaustion, or else utilization imposes an undesirable environmental hazard or economic handicap. Nevertheless, the world demand for energy is expected to increase everywhere as a result of the increasing population of our planet and as a consequence of the universal aspiration for an ever higher material standard of living. The ubiquitous need for more energy has currently stimulated the proposal of several unconventional technologies as possible remedies for this demand. Some of these proposals are merely further extensions of currently existent systems, e.g., fast breeder reactors, geothermal heat, or in situ coal gasification. Such schemes certainly contain their share of challenges and uncertainty, although much of the initial research and development is already completed. There are, however, other technological approaches which are even newer but which hold equal, if not superior, promise for mitigating the so-called energy crisis. In the latter category, controlled thermonuclear reactors (CTR) are high on the list.

As a source of energy, controlled thermonuclear fusion has many attractive features. It constitutes a substantially reduced radioactive hazard

compared to conventional or fast breeder reactors and is generally more acceptable from an ecological viewpoint. The economics for fuel production are extremely favorable, and the potentially usable fuels are themselves of almost unlimited abundance. However, we have scarcely begun to solve the problems attendant upon a CTR; in fact, it is almost certain that many serious problems remain unidentified. Nevertheless, in recent years we have made considerable progress toward solving the problem of plasma confinement, and the next major research phase in the development of this complex machine is beginning to emerge. This phase, or area of activity, bears the general title of "CTR Technology" as distinct from CTR Plasma Physics. Of the many problems and areas for research which have been specified thus far, we will deal here solely with one resulting from radiation damage incurred by the inner wall of a CTR as a result of bombardment by high-energy neutrons.

It would not be appropriate to review here all the various confinement geometries which are now being investigated at various laboratories around the world, since this has been done elsewhere.¹ The essential features with regard to radiation damage which appear almost regardless of design are shown schematically in Fig. 1. A very hot plasma is confined by means of magnetic fields which are produced by superconducting coils. An exception to this general design is laser fusion, which nevertheless will produce most of the same materials problems. Regardless of the confinement geometry, it is generally agreed that the following nuclear reactions are the most popular candidates for a CTR:

	Reaction	ΔE (MeV)	Threshold (keV)
(1a)	$2D \longrightarrow T + H$	3.25	50
(1b)	$2D \longrightarrow {}^3He + n^0$	4.0	50
(2)	$T + D \longrightarrow {}^4He + n^0$	17.6	10
(3)	${}^3He + D \longrightarrow {}^4He + H$	18.3	180

For a variety of reasons, but mainly because of its large cross section (see Fig. 2) low ignition temperature and large energy yield, reaction (2) is now the most highly favored of the lot.

The tritium necessary to sustain the reaction is produced by reacting the neutrons with lithium according to the following reactions:

Fortunately, the naturally occurring relative isotopic abundance, ${}^6Li/{}^7Li = 0.08$, is very nearly optimum for production of T. 7Li has its maximum neutron cross section at 2.8 MeV, whereas 6Li has its maximum at thermal energy. Thus a molten Li moderator serves as the primary heat exchange medium while also serving as a fertile fuel for the production of tritium.

The temperature of the inner wall, which interfaces with the plasma, is conceived to be at a temperature of $\sim 600^\circ \pm 200^\circ C$. Compared to the temperature of the plasma itself, $\sim 10^8^\circ C$ (one hundred million), this must be regarded as somewhat on the cool side. The reason for sacrificing the obvious increase in Carnot efficiency to be derived from a higher temperature lies not in the physics or engineering complexities of the CTR, but in our

inability to use steam economically at significantly higher temperatures. By non-economic, we refer to the shortened lifetimes of generators operating much above 600°C.

The materials which we have elected to investigate are niobium, vanadium and molybdenum metals. Several feasibility studies have indicated that these materials offer the best selection of properties with respect to strength, activation, compatibility, etc. Unfortunately, from the engineering point of view, these elements and their alloys have received very little attention from those doing basic research in radiation damage. It should also be noted that relatively little scientific research into the nature of neutron-induced radiation damage has been completed thus far. Our ignorance is therefore compounded by our choice of materials as well as our choice of radiation. Because of this overall lack of information, we have embarked on a fundamental investigation of the interaction of energetic neutrons with metals and alloys as distinct from an engineering study. Another justification for basic research stems from our inability to achieve the very high fluxes of high-energy neutrons anticipated for controlled thermonuclear reactors. Several estimates² have been made of CTR fluxes which range from $\sim 3 \times 10^{14}$ to 10^{15} n/cm²/sec, of which ~20% will consist of 14-MeV neutrons. There does not exist today a sustained source of 14-MeV neutrons which can meet even the lowest of these estimates. Consequently, we must rely upon simulation rather than direct measurement, which in turn requires a fundamental understanding of the physics.

Neutron bombardment can displace atoms in a solid matrix by two different mechanisms, viz, elastic and nonelastic. The former is the simplest and most straightforward whereby the neutron collides with the nucleus and

the resultant displacement conserves both energy and momentum. The nonelastic process itself can be divided into two mechanisms: one which leaves the nucleus unchanged, e.g., $\text{Nb}(n',n)\text{Nb}$ and one which does not, e.g., $^{93}\text{Nb}(n,\beta^-)^{94}\text{Zr}$. Energetic neutrons can give rise to a complete spectrum of nuclear reactions which include (n,α) and (n,γ) as well as fission. Further atomic displacements are then made possible via the interaction of the various radioactive emanations with the host atoms. The neutron has no interaction with electrons and hence loses its kinetic energy only as the result of nuclear collisions. The cross section for such collisions in nonfissile material is of the order of 1 barn (10^{-24} cm^2), and the mean free path between collisions is of the order of several centimeters.

Even at very high energies, the neutron behaves classically in elastic collisions, and a straightforward consideration of the conservation of momentum and energy permits one to calculate the energy transferred. For a mono-energetic beam of neutrons, of kinetic energy E_1 , the energy transferred, E_2 , is given by

$$E_2 = \frac{4M_1M_2}{(M_1 + M_2)^2} E_1 \sin^2\left(\frac{\theta}{2}\right) = \frac{4M_2}{(1 + M_2)^2} E_1 \sin^2\left(\frac{\theta}{2}\right), \quad (1)$$

where θ is the angle between the direction of the incident neutron and the displacement direction of the recoiling atom. For example, a 14-MeV neutron colliding headon with a niobium atom would transfer as its maximum amount of energy

$$E_2 = \frac{4 \times 93 \times 14}{(94)^2} = 0.6 \text{ MeV}. \quad (2)$$

Assuming the scattering to be isotropic, we can estimate an average value of $\bar{E}_2 = 0.300$ MeV. This estimate ignores the effect of nonelastic collisions which will lower somewhat the average energy of the primary knock-on (PKO). Taking $\bar{E}_2 = 150$ keV, we can estimate the total number of knock-ons, n , according to the simple formula of Kitchen and Pease,

$$n = \bar{E}/2E_d = 150/2 \times 0.025 = 3 \times 10^3. \quad (3)$$

This is also somewhat of an overestimation as it neglects that fraction of the energy $\bar{E}_2$ which is dissipated by simple ionization and is thus not available for causing atomic displacements. More sophisticated models include corrections for this as well as the energy dependence of the displacement efficiency. Nevertheless, the elementary theory is sufficient to demonstrate the relationship between the neutron energy and the total number of displacements. What has not been shown is the relationship between the ultimate character of the damage and the energy spectrum of the PKO's. It is intuitively obvious that high-energy PKO's will produce regions of highly concentrated damage in contrast to less energetic events which will create more-or-less isolated Frenkel pairs. The ability of the crystal defects produced by radiation damage to survive the annealing which is concurrent with the irradiation will depend upon their initial geometric disposition; the structural properties of the material will in large measure be governed by the character and concentration of the surviving defects. Hence, we have established the practical need for investigating radiation damage caused by 14-MeV neutrons.

Our present source of 14-MeV neutrons is much weaker than anticipated for a CTR. Nevertheless, when combined with the results of high-flux fission neutrons such as produced by EBR-II (Experimental Breeder Reactor No. II), it will provide an adequate basis for simulation. The source we refer to is the Rotating Target Neutron Source (RTNS) located at the Lawrence Livermore Laboratory (LLL).³ The RTNS, currently operational, delivers a peak fluence of approximately 3×10^{12} n/sec. Approximately half this amount can be obtained as useful flux on the experimental specimen. These neutrons are produced by impinging a 400-keV, 5-10 mA beam of D^+ upon a titanium tritide target ($\sim TiT_{1.2}$). The target is $3-5 \text{ mg/cm}^2$ thick ($\sim 10 \mu$), supported on a copper backing which is cooled by flowing water over the outer surface; the entire assembly is shown in Fig. 3. The water-cooled target is rotated at a velocity of 1100 rpm which avoids the problem of local heating while simultaneously maximizing the lifetime of the target. The neutron flux is determined by means of proton recoil telescopes which are in turn calibrated by neutron activation analysis. Reference 3 contains a more detailed description of the RTNS.

Recent calculations by C. M. Logan⁴ imply that 16-MeV protons create a displacement cascade in which the energy distribution is very nearly identical to that calculated for 14-MeV neutrons. His results are shown in Fig. 4, and the interested reader is referred to Ref. 4 for complete details of the calculations. The preliminary experiments to test the validity of these results are discussed next. The possibility of using protons to simulate high-energy neutrons is indeed tantalizing, for one can easily

obtain particle fluxes comparable to a CTR. The major drawbacks of proton experiments are first, the very low penetration distances (~ 100 - 200μ) and second, the experimental difficulty in removing the heat efficiently so as to maintain a reasonable target temperature.

The initial experiments have consisted of comparing the appearance of various metals which have been irradiated with fission neutrons at ambient temperatures with others irradiated by the RTNS. The dosages are comparable, although an accurate knowledge of the total cross section as a function of neutron energy makes it impossible to obtain an accurately normalized comparison. The fission neutrons are obtained in the Livermore Pool-Type Reactor (LPTR) in which the ambient temperature is $\sim 45^\circ\text{C}$ and the total flux is $\sim 10^{13}$ n/cm²/sec. The irradiation of high-purity Cu⁵ makes clear the difference between damage caused by 14-MeV neutrons and those of lower energy, as shown in Fig. 5. The visible radiation damage consists of clusters of vacancies or self-interstitials produced by irradiation to $\sim 3 \times 10^{16}$ particles/cm² of 18-MeV protons, 14-MeV neutrons and fission neutrons. These clusters lie on the $\{111\}$ planes and are in effect stacking faults with Burgers vectors $a/3\langle 111 \rangle$. The difference in the concentration and size of the damage clusters between the LPTR specimen and the other two is quite obvious. In the case of both the 14-MeV n⁰ and p⁺ irradiated specimens, the result appears to be qualitatively similar to a specimen having received a much larger dose of fission spectrum neutrons. Figure 6 records a semi-quantitative measurement of the cluster size and density for all three specimens and serves to emphasize the similarity of the 18-MeV p⁺ and 14-MeV neutrons.

The most direct method of simulating the PKO spectrum is by self-ion bombardment. We have not actively pursued such a program at LLL; nevertheless, for the sake of completeness we include a photograph (Fig. 7) of a 304 stainless steel foil bombarded by 5-MeV Ni^{++} ions.* One can easily see the high concentration of small voids resulting from the large number of displacements. (Each atom displaced ~ 50 times.) Similar microstructures have been produced by high neutron fluences, and this phenomenon will be discussed in another paper at this Conference. (See Problem Areas in the First Wall Materials of a Controlled Thermonuclear Reactor by A. C. Damask.)

The profound effect of impurities upon the damage structure can scarcely be overemphasized. For example, the addition to vanadium of 3-20% titanium effectively suppresses the formation of voids.^{6,7} Figure 7 shows Nb doped to various levels with oxygen and then irradiated in the Argonne CP-5 Reactor to various total fluences.[†]

Radiation-enhanced diffusion offers an indirect method for observing the effect of varying dose and neutron energy. It can also yield qualitative information about the stability of phases or compositional, short-range order in radiation fields. We have chosen to observe the change in the magnetic order of Au-Fe alloys by means of the Mössbauer effect as a function of the irradiation variables. A previous investigation⁸ established that the temperature at which magnetic order begins and the temperature

*We wish to thank Mr. P.B. Mohr of LLL for permission to use these results.

†We wish to thank Dr. B.A. Loomis of ANL for permission to use these results.

dependence of the magnetic hyperfine splitting are quite sensitive functions of the short-range compositional order. The order was varied by simply annealing in the two-phase region a single-phase, α -solid solution of Au-17% Fe. The annealing temperature was high enough to allow atomic migration, and the net driving force is in the direction leading to the separation of two phases, one Fe-rich and the other Au-rich. The anneal was interrupted before the actual precipitation of a second phase, but only after considerable clustering into Au-rich and Fe-rich regions had occurred. The increased temperature at which magnetic order starts as a result of such clustering is illustrated in Fig. 8. Note, however, that irradiation by fission neutrons causes an even more pronounced segregation as evidenced by the much better developed hyperfine spectrum. Additional evidence for the efficacy of neutrons in aiding the production of solute clusters is shown in Fig. 9. Here, the measured magnitude of the splitting, $H(T)$, is assumed to be a measure of the departure from random order, and it is obvious that $\sim 10^{16}$ n/cm² at $\sim 45^\circ\text{C}$ induces a greater degree of clustering than annealing for several days in the two-phase region.

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FIGURE CAPTIONS

1. Schematic showing the essential features of a CTR.
2. Plots of fusion reaction rate parameters $\langle\sigma v\rangle$ for d-t, d- ^3He , d-d (total), and p- ^6Li reactions as obtained by averaging over a Maxwellian distribution; 1 keV kinetic temperature $\approx 1.16 \times 10^7$ deg Kelvin (taken from Ref. 1, courtesy of R. F. Post).
3. Schematic of the Rotating Target Neutron Source (taken from Ref. 3, courtesy of R. Booth).
4. Calculated total PKO spectra in Nb. Energy spectra of recoiling Nb atoms from collisions with 14.1-MeV neutrons and 16.4-MeV protons (taken from Ref. 4, courtesy of C. M. Logan).
5. Transmission electron micrographs of radiation damage structure in high-purity Cu. Kinematic diffraction contrast (taken from Ref. 5, courtesy of J. B. Mitchell).
6. Voids in 304 stainless steel as a result of bombardment by 5 MeV Ni^{++} ions (courtesy of F. B. Mohr).
7. Radiation damage in Nb doped to various concentrations with O_2 and irradiated with fission spectrum neutrons (courtesy of B. A. Loomis).
8. Comparison of the Mössbauer spectra of a Au-17% Fe alloy which has been (a) quenched from the single-phase region, (b) quenched, then annealed for several days in the two-phase region, and (c) quenched, then irradiated to $\sim 10^{16}$ nvt with reactor spectrum neutrons.
9. The magnitude of the magnetic hyperfine splitting of Au-17% Fe alloy shown as a function of various degrees of short range order (see text for explanation). Note the greater values of H at equivalent temperatures for the irradiated specimen.

CONCEPTUAL 12,000 MW_t FUSION REACTOR

BASED ON N.C. CHRISTOFILOS' ASTRON PLASMA CONTAINMENT SCHEME

Borg - Fig. 2

Borg - Fig. 3

Borg - Fig. 4

75-12

100 000 X

 $Z \approx [310]$ $g \approx [002]$ $t \approx 2100 \text{ \AA}$

Fission Neutrons - LPTR

7U-1

100 000 X

 $Z \approx [510]$ $g \approx [002]$ $t \approx 1780 \text{ \AA}$

14-MeV Neutrons - RTNS

75-23

100 000 X

 $Z \approx [100]$ $g \approx [002]$ $t \approx 1750 \text{ \AA}$

16-MeV Protons

Borg - Fig. 6

Borg - Fig. 7

Borg - Fig. 8

Borg - Fig. 9