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SPALLATION RADIATION DAMAGE AND DOSIMETRY FOR ACCELERATOR TRANSMUTATION OF WASTE APPLICATIONS

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ABSTRACT: Proposals are currently being made for systems to treat radioactive waste based on the use of accelerator-driven neutron sources. A linear proton accelerator with energies as high as 1600 MeV and currents up to 250 ma are anticipated for the driver. The neutron fluxes may reach up to 10^{20} neutrons/m²s as generated by the spallation reactions that occur when the protons strike target materials. Calculations are described to determine radiation fluxes and flux spectra inherent in such systems and to estimate likely radiation effects on system components. The calculations use LAHET, a Monte Carlo high-energy transport code, and MCNP, a generalized-geometry, coupled neutron-photon Monte Carlo transport code. Cross sections for displacement and helium production are presented for spallation neutrons of energies from 21 MeV to 1600 MeV for Inconel 718 (Ni plus 18.5, 18.5, 5.1, and 3 wt% of Cr, Fe, Nb, and Mo, respectively), an alloy that is used for the proton beam entry window in several accelerators. In addition, results for this alloy are presented for the primary knocked-on atom (PKA) spectrum and the transmutation yield for 1600 MeV incident neutrons.

KEYWORDS: accelerator transmutation of waste (ATW), displacement production, helium production, Inconel 718, LAHET, MCNP, PKA spectrum, proton accelerator, radiation damage, and transmutation production

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Spallation neutrons are produced when protons of energies higher than about 100 MeV strike materials in their path. At these energies the DeBroglie wavelengths of the protons are so short that they interact separately with individual nucleons, causing the nucleons (neutrons and protons) to be ejected or spalled out of the nucleus. The resulting spallation neutron spectrum consists of a degraded fission spectrum with a high-energy tail extending up to the energy of the incident proton. Proton-accelerator-driven spallation neutron sources are used as the basis for neutron diffraction and scattering facilities, such as the Los Alamos Neutron Scattering Center (LANSCE) [1] at the Los Alamos National Laboratory (LANL), the Intense Pulsed Neutron Source (IPNS) [2] at the Argonne National Laboratory, and SINQ at the Paul Scherrer Institute (PSI) in Switzerland [3]. In these cases, D₂O moderation is used to thermalize the spallation neutrons. Alternatively, the unmoderated spallation neutrons may be used for materials irradiation studies, as for example at the Los Alamos Spallation Radiation Effects Facility (LASREF) [4,5], which is driven by 800 MeV protons at a current of about 1 ma. The resulting spallation neutron flux at the copper beam stop is about 5×10^{17} neutrons/m²s, and calculations have shown [6] that the flux can be increased by a factor of about 10 by removing extraneous materials in the neighborhood of the beam stop, changing the beam stop material from copper to tungsten, and decreasing the beam diameter from the current value at the Los Alamos Meson Physics Facility (LAMPF) of 0.105 m to 0.052 m.

Accelerator-driven neutron sources have lately received attention in connection with studies of accelerator transmutation of waste (ATW) concepts, such as those presented by investigators at Los Alamos National Laboratory [7-9] and Brookhaven National Laboratory [10,11]. The basic idea is to produce spallation neutrons by bombarding appropriate high-atomic-weight target materials in a high-current proton accelerator. The spallation neutrons are then moderated and nuclear waste materials in solution or in a slurry are circulated through the high-flux thermal-neutron region that is thus produced. This increases the immediate radioactivity of the nuclear waste, but greatly decreases its half-life. The proton current and energy for the ATW accelerator are projected to be in the ranges 5-250 ma and 800-1600 MeV, respectively. These parameters are considered to be reasonable scale-ups of the 1 ma - 800 MeV LAMPF proton accelerator in the light of advances achieved in accelerator technology since LAMPF began operating about two decades ago [12]. In the context of considerations of the feasibility of accelerator production of tritium (APT), a joint Los Alamos - Brookhaven study group developed the design concept for a 200 mA - 1600 MeV linear accelerator. The design was judged to be technically sound by the Department of Energy (DOE) Energy Research Advisory Board [13]. A key feature of an ATW system operating at the power levels indicated above is that the heat produced permits the production of sufficient electric power to not only drive the accelerator but, in addition, to feed significant amounts of power to the electric grid [9]. Thus, the ATW system serves to reduce the long-term radioactivity of nuclear wastes and, at the same time, produce net electric power.

The number of spallation neutrons produced per incident accelerator proton increases with increasing atomic number of the target material. Thus, heavy target materials are preferred. Target design options fall into two broad categories: (1) liquid targets with flowing Pb-Bi eutectic or liquid Pb, and (2) solid pebble-bed targets, using W or Pb pebbles or perhaps a combined system with W pebbles in a central cylinder surrounded by an annulus of Pb pebbles. The W and Pb pebbles would be cooled by helium and water, respectively. Additional design concepts have described water-cooled, clad and unclad, W and Pb rods and plates. Neutron flux calculations for a 50 ma - 1600 MeV spallation source with a liquid Pb-Bi target indicate a peak flux at the target

container wall approaching 10^{20} neutrons per m^2 s with energies extending up to 1600 MeV [14]. In radiation environments of this type, there can, of course, be significant radiation damage to the materials used for beam access windows, container alloys, and other structural materials critical to the operation of the ATW systems. In a recent report [15], a general discussion is given of the factors affecting the selection of materials for ATW systems. In the present paper, we describe the calculation and measurement of the spallation neutron flux and flux spectrum anticipated for ATW systems. In addition, radiation damage characteristics are described as calculated for a typical accelerator beam entry material, Inconel 718 (Ni plus 19, 18.5, 5.1, and 3 wt% of Cr, Fe, Nb, and Mo, respectively). These characteristics include displacement, helium production, and transmutation production cross sections and PKA spectrum.

SPALLATION NEUTRON DOSIMETRY

Measurements of the spallation neutron flux and spectrum have been conducted to a limited extent at LASREF using foil activation dosimetry techniques [16-19]. A rabbit system was used that permitted aluminum capsules to be driven by helium gas through tubes to and from the irradiation locations. Four rabbit tubes were situated at distances from 0.12 to 0.38 m from the proton beam centerline, each of which could accommodate five aluminum capsules containing Fe, Co, Ni, Ti, V, Sc, Al, Zn, Au, and Cu foils. The foils were irradiated for periods of 3-62 h. Upon removal from the accelerator, the radioactivities of the foils were measured using a calibrated high-purity germanium crystal counter. The SPECANL computer code [20] was employed to analyze gamma ray spectra, and the neutron spectrum was evaluated with the STAY'SL code [21]. In STAY'SL, the best least-squares spectrum that produces the measured radioactivities is determined, starting with a particular input spectrum, which was calculated in the manner described in the next section. A comparison of the measured neutron flux versus neutron energy with the calculated one provided an indication of the experimental accuracy. The comparison was best for neutron energies above about 0.01 MeV. For lower neutron energies, the measured neutron fluxes were lower than the calculated ones.

SPALLATION NEUTRON FLUX AND SPECTRUM CALCULATIONS

The principal computer codes employed for the calculation of spallation neutron flux and spectrum are the Los Alamos High Energy Transport Code (LAHET) [22] and the Monte Carlo Code for the Transport of Neutrons and Photons (MCNP) [23]. LAHET is the LANL version of the High Energy Transport Code (HETC), which was originally developed at the Oak Ridge National Laboratory (ORNL) [24] to describe the nuclear products resulting from inelastic proton-nucleus collisions. LAHET and HETC use the two-stage spallation model, as originally suggested by Serber [25], in which spallation reactions consist of an intranuclear cascade, followed by evaporation or fission as the excited nucleus attempts to return to its ground state. LAHET calculates the direction and energy of emitted cascade and evaporation particles and the mass and energy of the recoiling residual nuclei, as well as the masses and energies of recoiling fission products. It also transports the particles through the geometry until they degrade down to their cutoff energies, which is 20 MeV for neutrons. A neutron leaving a reaction with less than 20 MeV has its kinetic parameters recorded in a file for subsequent transport by MCNP using ENDF/B-based neutron cross section libraries. Thus, LAHET and MCNP operate as a coupled system. Tallies are then available from MCNP, which provide the desired flux and spectrum information.

In the present calculations, the incident proton energy was taken to be 1600 MeV. The target was assumed to be a homogeneous mixture of tungsten and water, where the water is considered to function as coolant. The volume fraction occupied by the tungsten was 0.85. The target radius was set at 0.25 m, consistent with the information in [9]. Heavy water surrounded the target to serve as moderator. The calculations were performed on an HP 9000/735 workstation. The number of incident protons was 50,000, which resulted in over 10^6 neutrons with energies below 20 MeV to be transported in MCNP. About 3 h of CPU time was required per Monte Carlo run. The history files generated comprised about one gigabyte. The neutron flux and spectrum were tallied at two locations: at the surface of the beam stop (i.e., at 0.25 m from the axis of the proton beam) and at 0.40 m from the beam axis. The calculated spallation neutron flux and spectrum for the two locations are shown in Fig. 1. The total flux of neutrons of all energies at the 0.25 m location was 1.3×10^{20} neutrons per $m^2 \cdot s$. It should be realized, however, that our simulation of an ATW system lacked a number of features that might be present in a real ATW facility. For example, our simulation did not include fissioning actinides in the blanket. In addition, we did not surround the blanket with reflector materials, which might be present in an actual ATW system.

Fig. 1. Spallation neutron flux at 0.25 m (upper curve) and 0.40 m (lower curve) from the axis for 1600 MeV protons on a homogeneous mixture of tungsten and water surrounded by D₂O.

Cappiello [14] presented neutron flux calculations for a four-region ATW facility. The target (Region I) was liquid Pb-Bi eutectic surrounded by an inner D₂O-Tc⁹⁹ slurry (Region II), a D₂O-Np slurry (Region III), and an outer D₂O-Tc⁹⁹ slurry. The calculations assumed a 50 ma - 1600 MeV proton beam. The total neutron flux at the outer edge of the target was about 8×10^{19} neutrons/ $m^2 \cdot s$. Similar results were reported by LaBauve, Cappiello, and Perry [26].

RADIATION DAMAGE CALCULATIONS

Displacement Production and PKA Spectra

Earlier calculations of displacement production in Cu due to 800 MeV protons [27] and due to the resulting spallation neutrons [28] used the code known as VNMTTC, which is based on the Nucleon Meson Transport Code (NMTC) of Coleman and Armstrong [29]. In the present work, LAHET was used to calculate the displacement cross section for the Inconel 718 alloy (considered to be a cylinder 0.5 m in radius and 0.01 m thick) for incident neutrons from 21 to 1600 MeV. Each run consisted of 100,000 incident neutrons. The LAHET output provides the average damage energy per incident neutron, T_{dam} , calculated in accord with the Lindhard model [30] for the partitioning of atom recoil energies between nuclei (which produces displacements) and electrons (which does not produce displacements). The thin-sample displacement cross section is then given by

$$\sigma_d = \frac{\beta T_{dam}}{2T_d N_V x}$$

where T_d is the threshold displacement energy (taken here to be 40 eV), β is 0.8, N_V is the atom density, and x is the sample thickness, 0.01 m. N_V was calculated from the density of 8.22×10^4 g/m², as given in [31].

Figure 2 shows that the displacement cross section increases rapidly for incident neutron energies above 300 MeV and then levels off at about 3200 barns for energies above 800 MeV. The damage energy cross section corresponding to 3200 barns is 320 barns keV. By contrast, the earlier calculations for 800 MeV neutrons on Cu gave a damage energy cross section of about 400 barns keV [15,28].

Fig. 2. Displacement cross section versus incident neutron energy for Inconel 718 ($T_d = 40$ eV).

LAHET also provides the number of recoils per incident neutron, n/n' , falling into specified energy bins, where the energy refers to the raw recoil energy (atom recoil energy that is partitioned among both nuclei and electrons) or to the damage energy (energy delivered only to nuclei, as explained above). The bin structure specified starts at 1 eV, and each higher bin boundary energy is twice the previous one. The n/n' was divided by the bin width, and this was divided by N_{yx} to give the differential scattering cross section with respect to the energy T transferred to the atom, σ_T , i.e., to give the PKA spectrum. The PKA spectrum with respect to damage energy is shown in Fig. 3 for incident neutrons of energy 1600 MeV. The most probable damage energy is seen to be about 0.1 MeV, corresponding to the peak of the curve. The LAHET output for this case gives an average damage energy of 0.27 MeV.

Fig. 3. PKA spectrum with respect to damage energy for 1600 MeV neutrons on Inconel 718.

Helium Production

Helium production is an important consideration because of the tendency of helium to aggregate into bubbles at grain boundaries, thus promoting high-temperature grain boundary cracking. The LAHET output gives the number of helium atoms produced per incident neutron, and this dividing by N_{yx} again gives the thin-sample production cross section, which is shown in Fig. 4 as a function of the incident neutron energy. The helium production cross section is seen to increase monotonically with increasing neutron energy, reaching values approaching 0.5 barns at 1600 MeV.

Transmutation Production

Transmutation yield in irradiated ATW materials needs to be determined, of course, because transmutation products constitute impurities that may have a profound effect on properties. LAHET has an option, Detailed Residual Masses, that gives the numbers of nuclei per incident neutron with specified proton and neutron numbers remaining after the spallation process is completed. The transmutation production cross section may then be calculated in a manner similar to that

described above. Fig. 5 shows for Inconel 718 and for incident neutron energy of 1600 MeV the transmutation production cross section versus atomic number, Z , of the transmutation product, for all isotopes with the indicated Z . The transmutation yield tends to be high for Z numbers below and near that of the target. Since the major elements in Inconel 718 are Ni ($Z=28$), Fe ($Z=26$), and Cr ($Z=24$), the transmutation yield is peaked in the range of about $Z=22-27$. It is to be noted, however, that the yield extends all the way down to about $Z=6$, so that minor amounts of particularly injurious impurities might well be produced.

Fig. 4. Helium production cross section versus incident neutron energy for Inconel 718.

Fig. 5. Transmutation production cross section versus atomic number of the transmutation product for Inconel 718 irradiated with 1600 MeV neutrons.

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