

Radiation Damage to Heavy Target Materials Induced by High Energy Proton and Electron Beams

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Abstract

Nuclear waste transmutation is an important concept in the development of an advanced fuel cycle. Along with reactor-based approaches, waste transmutation using accelerators is being investigated. Currently, the most widely accepted concept for accelerator-based transmutation is with the use of a high-current proton accelerator. There has also been some interest in investigating the efficacy of using electron accelerators for this purpose. In contrast to high-current proton accelerators, electron accelerators use well known technology, that is been used in a variety of other applications. With the availability of high energy electron beams, electron accelerators are capable of producing a high level neutron flux.

One of the concerns with the use of high energy particles for neutron generation/waste transmutation is radiation damage to targets and surrounding materials. The proton beam induces spallation reactions in targets which typically results significant radiation damage. Having a much smaller particle mass, electrons would be expected to cause much less radiation damage. With an electron beam, however, the resulting Bremsstrahlung photons will also cause radiation damage.

In this study, radiation damage to materials was analyzed both for proton and electron accelerator beams. Materials selected for the study were used target lead and uranium. The MCNP-X code was used for the supporting calculations. Targets were set up in a cylindrical geometry. Details of flux and spectra distributions in the target were characterized for protons and electrons, and for photons and neutrons. With the availability of electron, proton and neutron fluxes and relevant reaction cross sections, the radiation damage products and displacements were calculated by folding the proton or neutron flux data into the corresponding cross section with the source beam assumed at 1mA and 1000MeV. Radiation damage was calculated for 10,000 hours of irradiation. Results indicate that radiation induced displacement typically is more than 30 times greater with the proton accelerator beam than with the electron accelerator beam.

Introduction of Radiation Damage Production to Target

Radiation damage to material is generally defined as the alteration of material properties arising from exposure to ionizing radiation. Harmful changes in the properties of liquids, gases, and solids, caused by interaction with nuclear radiations. The damage caused by the removal of atoms from a solid material when elementary particles, such as those associated with radioactivity, collide with it. Radiation damage is an important consideration in the design of nuclear reactors, where radiation levels are high.

The initial event in the damaging of a crystal lattice by high energy radiation is the sudden transfer of a rather large amount of kinetic energy to a single atom. The energized atom then ploughs through the lattice knocking other atoms from their sites and leaving a damaged region behind. From a theoretical standpoint this damaging event is a complex many-body problem, it has been treated in the past by making drastic approximations [1]. Generally it has

been considered as a cascade of independent, two-body collision between knock-on atoms and stationary atoms.

The radiation inside a reactor generally consists of fast and thermal neutrons, γ -rays and a relatively few Beta particles. The proportions of these radiations vary not only between one reactor and another, but also from one part of a reactor to another. In the majority of cases, however, the overwhelming majority of displaced atoms are produced by fast neutron bombardment [2].

Radiation production of displacement of atom

Kinchin and Pease (1955) had described the displacement of atoms in solids by radiation including electrons, gamma and neutrons radiation effect. The fast neutrons used in radiation work are usually produced by fission. The cross section for producing displacement is the total neutron cross section. Electrons radiation displacement is predominant by Coulomb interaction with nuclei. The gamma ray production of displace received very little

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attention due to high threshold radiation energy and low radiation production cross section. Radiation generally produces groups of displaced atoms after a time t and temperature T . The total number of displaced atoms is an accumulated effect and is roughly through constant throughout the radiation. Threshold radiation energies were considered for various types of radiation, as shown in Table 1. The threshold energy was selected at 25 eV for both of the two targets [3]

Table 1: Threshold Radiation Energy for Displacement [2]

Threshold Radiation Energy for Displacement				
Atomic weight of stationary atoms	10	50	100	200
Neutrons, protons (eV)	76	325	638	1263
Electrons, r-rays (MeV)	0.1	0.41	0.68	1.1
Fission fragments (eV)	85	30	25	27

In the proton-based target, the proton and neutron radiation damage are taken into account. In the electron-based target, electron and neutron radiation damage are considered. With the availability of electron, proton and neutron fluxes and the cross sections, the radiation damage products, displacements, are calculated by folding the proton or neutron flux into the corresponding cross section as described in:

$$R_p = \int_0^{\infty} \Phi'(E)X(E)dE$$

where R_p is the radiation product concentration, with unit as dpa

$X(E)$ is the cross section at energy E ,

$\Phi'(E)$ is the corresponding differential fluence $\Phi' = \Phi(E) / E$, where Φ is the differential neutron flux, $\Phi(E)$ is average neutron flux for given neutron energy E .

Unit: dpa, which is a measure of the amount of radiation damage generally in neutron-irradiated materials, for example, 10 dpa means each atom in the material has been displaced from its site within the structural lattice of the material an average of 10 times (due to interactions between the atoms and the energetic neutrons irradiating the material.)

This calculation method is widely adopted for the evaluation of displacement and proved to be useful. Meanwhile, the integration involves complicated calculation on cross section and differential incident flux.

Radiation Production Simulation

The calculations were made for two accelerator beams, proton beam and electron beam. Accelerator

beams of electron and proton are uniformly distributed area source with 1mA current. The beam radius was assumed as 1cm. The current was assumed at $6.25E+15$ #/s. Both proton and electron beam were transmitted through an infinite target. In the MCNPX [4] simulation model, the targets were set up with cylindrical geometry with particle beam upstream entrance at the cylinder bottom surface. A target material is selected with natural uranium, consisting of 99.3% U-238 and 0.7% U-235.

The calculation of radiation damage were performed using a standard method [5], which encloses the proton and neutron fluxes into corresponding cross sections for radiation products as displacement in a proton-based target; and encloses the electron, photon and neutron fluxes into corresponding cross sections for radiation products in an electron-based target. Processing of the calculation is described in the flow chart of Figure 1. It should be noted that the use of cross section sources is complicated as various cross section sources are involved with difficulty in checking the validity of the cross section sources.

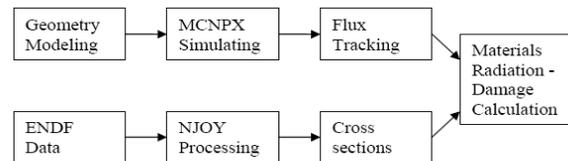


Figure 1: Flow chart of calculating the radiation damage production

In the flow chart, the first path is on MCNPX simulation in order to obtain the particle flux and spectrum. The second path is on the processing of cross sections [6]. Since the most of the evaluated cross section data are developed at room temperatures. Considerations need to be made to take into account the fact that in the accelerator bombard target, the temperature ranges from room temperature up to about 600 K. The temperature-dependant cross section was generated by processing NJOY [7].

Radiation Damage Production to Target Materials in Electron-based ADS

In the target of electron ADS, the interaction is mainly through of electron-induced Bremsstrahlung interactions, which produced high energy photons. The photon induces photonuclear interaction and produce photoneutrons. The flux distribution of all three particle flux distribution are plotted and shown in Figure 2. High energy electron hitting a target will result in the development of an "electron-gamma shower". A high energy electron produces a high energy photon (Bremsstrahlung). This photon then

produces electron-positron pairs and Compton electrons. These electrons then produce more photons by Bremsstrahlung. This continues until the collision losses dominate the radiative losses and the cascade or shower dies out. The Bremsstrahlung photons are forward directed. The angular distribution of photons

as a function of the energy of the incident electron favors forward movement for high energy electrons. The electrons and positrons generated by pair production are also forward directed; whose angular distribution is also a function of energy.

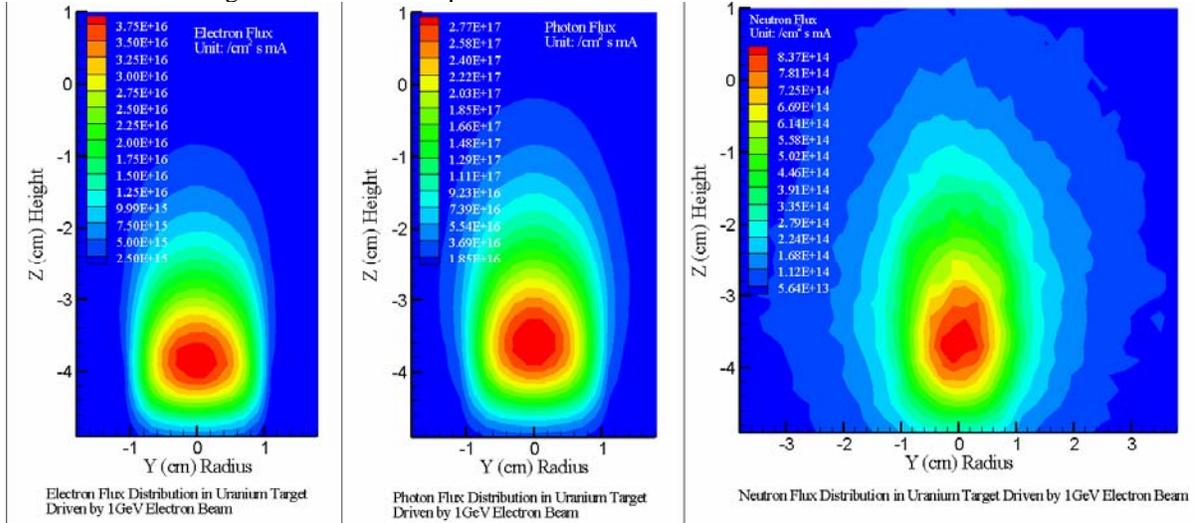


Figure 2: Electron, photon and photoneutron flux distribution in uranium target driven by 1GeV electron beam

In Figure 2, the red color stands for the peak flux with the peak electron flux as $3.75E+16 / \text{cm}^2 \text{ s mA}$, the peak photon flux as $2.77E+17 / \text{cm}^2 \text{ s mA}$ and the peak neutron flux as $8.37E+14 / \text{cm}^2 \text{ s mA}$. The magnitude of the peak electron flux is approximately one order of magnitude lower compared to that of the peak photon flux. But the peak flux level of the electrons is higher than the peak neutron flux level by about one order of magnitude. The location of the peak flux occurrence was observed approximately at the same location for all three particles, at about 1.5 cm upstream from the target entrance point.

The spectrum of particles, (electron, photon and neutron) is plotted in Figure 3. The tally/particle (Y-axis) in this figure stands for the average particle flux as $/\text{cm}^2$ per particle in the entire target volume. Considering the threshold energy involved, the electron energy was cut off if lower than 1MeV.

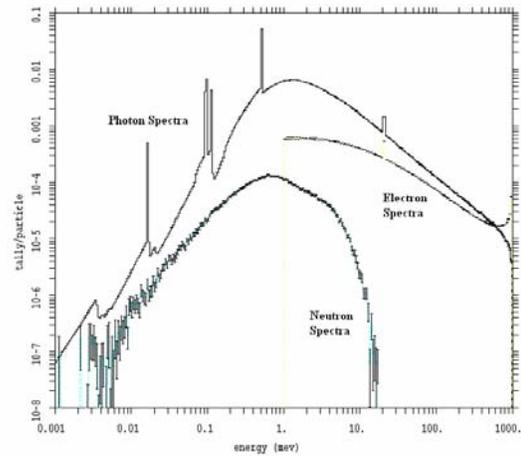


Figure 3: Electron, photon and photoneutron spectrum in uranium target driven by 1GeV electron beam

Radiation damage production was calculated after 10 Ah of exposures. The peak dpa from the electrons and neutrons was estimated as:

$$R_{pep} = 0.15 \pm 0.02 \text{ dpa by electron radiation}$$

where, R_{pep} stands for the peak radiation production by electron

$$R_{pnp} = 0.36 \pm 0.03 \text{ dpa by neutron radiation}$$

where, Rnp stands for the peak radiation production by neutron

The total radiation product in uranium target induced by 1GeV electron is about 0.51 +/- 0.05. That peak displacement occurs in the red zone in Figure 2. Outside the red zone, the radiation product is roughly proportional to the neutron flux.

Please note that the peak radiation damage production by photon is found to be much smaller by a couple of orders of magnitude compared to radiation damage by electrons, or neutrons. The contribution of photons to the total radiation damage production is insignificant and was ignored.

Radiation Damage Production to Target Materials in Proton-based ADS

In proton-based target, the neutrons are mainly produced in spallation reactions under the

bombardment of protons upon spallation target. The peak neutron flux occurred at the center of entrance of the target where the strongest reactions took place. The neutron flux decreased with increasing distance from the center of the peak neutron flux. The proton and neutron flux distribution inside the target are shown in Figure 4. A mesh tally of MCNPX was conducted along the beam direction, (i.e., Z-axis). For the target geometry as a cylinder, the flux distribution was plotted at the X=0 plane, (i.e., YZ plane). The red color stands for the peak flux in the figure. The peak flux was estimated to be 2.81E15 /cm² s mA and 2.54E+15 / cm² s mA for protons and neutrons, respectively. The magnitude of the peak flux of protons and neutrons were approximately equal. Neutrons were found to penetrate deeper and wider inside the target through proton-induced neutron generation and by fission.

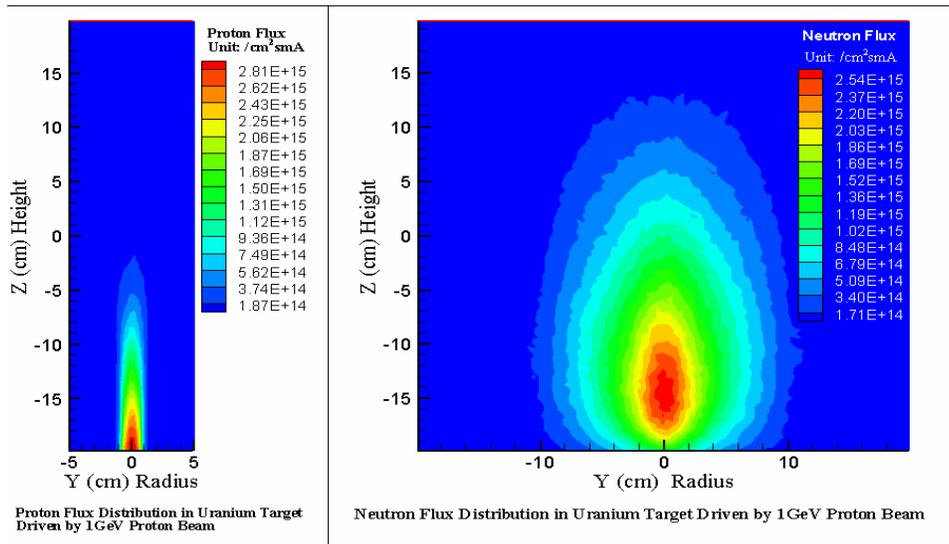


Figure 4: Proton and neutron flux distribution in uranium target driven by 1GeV proton beam

As the proton beam travels further into the uranium target, the proton flux drops quickly as more and more protons are lost through spallation reactions. Though there are secondary protons produced by spallation reactions, their contribution to the total proton flux is relatively small. Therefore, no apparent increase in proton flux is expected as observed in the figure.

The calculated proton and neutron spectrum is plotted in Figure 5. The tally/particle (Y-axis) in this figure stands for the average particle flux as /cm² per particle in the entire target volume. As observed, the majority of neutron energy was around several MeV, while the protons were at higher energy range (several hundred MeV up to 1 GeV).

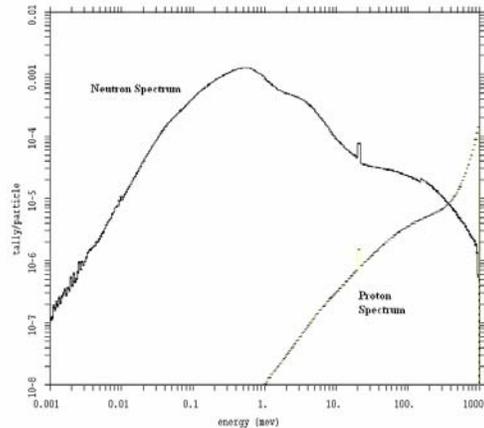


Figure 5: Proton and spallation-neutron spectrum Uranium target driven by 1GeV proton beam

Radiation damage production was calculated at 10Ah of exposure in the target from proton irradiation. The total displacement is the combination of proton and neutron radiation product, which is proportional to its corresponding flux.

Total radiation damage of the target entrance point, was estimated from the MCNPX calculations. The peak radiation damage production induced by proton and neutron was calculated as:

$$R_{ppp} = 9.41 \pm 0.34 \text{ dpa by proton irradiation}$$

where, R_{ppp} stands for the peak radiation production by proton

For neutron flux, the peak is along the Z-axis, at 3cm to 10cm away from the target entrance point.

The displacement by neutron radiation was estimated as:

$$R_{pnp} = 8.03 \pm 0.29 \text{ dpa by neutron radiation}$$

where, R_{pnp} stands for the peak radiation production by neutron

The total dpa reaches its maximum value of 17.4 dpa at 10cm upstream from the target entrance. Radiation damage calculation was performed on the uranium target induced by 1GeV protons and electrons separately. The flux distributions and energy spectrum of the particles were estimated by using MCNPX with the mesh tally method. Calculations of radiation damage were based on folding the neutron fluxes into corresponding cross sections for determining radiation products as displacement. The peak radiation product in proton-ADS was estimated to be higher than the peak radiation product in the electron-ADS by a factor of 34. In the proton-ADS, the protons and neutron have almost equally contributed to the total radiation product. In the electron-ADS, the neutrons have contributed almost twice more than the electron to the total radiation product.

Reference

- [1] F. Seitz and J. S. Koehler, in Solid-State Physics, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 2
- [2] Kinchin G.H., Pease R.S. The Displacement of Atoms in Solids by Radiation, Reports on Progress in Physics 18:1-51 1955
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Observation and conclusion

Radiation damage calculation is performed on uranium target induced by 1GeV proton and electron separately. The proton and neutron flux distribution and energy spectrum are provided in proton-based target. Meanwhile, the electron, photon and neutron flux distribution, and energy spectrum are obtained too simulated by MCNPX with mesh tally. The calculations of radiation damage discussed above are performed in a traditional manner, which folds the neutron fluxes into corresponding cross sections for radiation products as displacement. The total radiation damage product in uranium target and lead target driven by 1GeV proton and electron beam respective for 10Ah exposure are shown in Table 2.

Table 2: Combined Radiation Damage after 10,000 hour (~410 days), 1mA exposure

Radiation Damage Production by Accelerator Beam			
		Proton Beam	Electron Beam
U r a n i u m	dpa by Source Particle	9.41 +/- 0.34	0.15 +/- 0.02
	dpa by Neutron	8.03 +/- 0.29	0.36 +/- 0.03
	Total dpa	17.44 +/- 0.63	0.51 +/- 0.5
Proton Beam Electron Beam			
L e a d	dpa by Source Particle	8.95 +/- 0.33	0.14 +/- 0.02
	dpa by Neutron	10.11 +/- 0.36	0.45 +/- 0.04
	Total dpa	19.06 +/- 0.69	0.55 +/- 0.6

The peak radiation product in proton-based target is higher than the peak radiation product in electron-based target by a factor of 34. In proton-based target, the proton radiation product is approximately equally contributed to the total radiation product. In electron-based target, neutron radiation product are about 2 times contributes to the total radiation product. Proton beam travel to target much deeper and more concentrated on the beam direction, while electron beam travel into target in a short distance and then isotopic distributed.

- [5] Standard Practice for Neutron Radiation Damage Simulation by Charged-Particle Irradiation, E 521-96, Annual Book of ASTM Standards, Vol. 12.02, American Society for Testing and Materials, Philadelphia, 1996, pp. 1-20
- [6] Private communication with Dr. Wei Lu, North Carolina State University, for additional displacement cross section
- [7] NJOY Nuclear Data Processing System, Version 99, <http://t2.lanl.gov/codes/njoy99/>, Los Alamos National Laboratory, Los Alamos, NM, 2000