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Application of the Monte Carlo method to estimate doses due to neutron activation of different materials in a nuclear reactor

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A R T I C L E I N F O

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A B S T R A C T

All materials exposed to some neutron flux can be activated independently of the kind of the neutron source. In this study, a nuclear reactor has been considered as neutron source. In particular, the activation of control rods in a BWR is studied to obtain the doses produced around the storage pool for irradiated fuel of the plant when control rods are withdrawn from the reactor and installed into this pool. It is very important to calculate these doses because they can affect to plant workers in the area. The MCNP code based on the Monte Carlo method has been applied to simulate activation reactions produced in the control rods inserted into the reactor. Obtained activities are introduced as input into another MC model to estimate doses produced by them. The comparison of simulation results with experimental measurements allows the validation of developed models. The developed MC models have been also applied to simulate the activation of other materials, such as components of a stainless steel sample introduced into a training reactors. These models, once validated, can be applied to other situations and materials where a neutron flux can be found, not only nuclear reactors. For instance, activation analysis with an Am-Be source, neutrography techniques in both medical applications and non-destructive analysis of materials, civil engineering applications using a Troxler, analysis of materials in decommissioning of nuclear power plants, etc.

1. Introduction

All materials exposed to some neutron flux can be activated independently of the kind of the neutron source. There are four types of neutron sources: nuclear reactions with particles that usually are (α, n); nuclear reactions with high energy photons, (γ, n); fission reactions in nuclear reactors; and accelerators. In this study, a nuclear reactor has been considered as neutron source.

Activation reactions can be simulated with the MC method (e. g. MCNP5) and the number of reactions calculated converted into activity. Activated materials can produce a dose around them. This dose is a potential risk for people staying in the surrounding area. Therefore, it is necessary to assess the activity generated and the dose produced.

In particular, materials present in the core of a nuclear reactor become activated by neutron irradiation. When activated materials are withdrawn from the reactor, a dose is produced around them. It is, of course, a risk for workers that should be estimated.

The origin of this study was the analysis of the activation of control rods in a BWR to obtain the doses produced around the storage pool for irradiated fuel of the plant when control rods are withdrawn from the reactor and installed into this pool. It is very important to calculate these doses because they can affect to plant workers in the area.

The MCNP5 code (X-5 Monte Carlo Team, 2005) based on the Monte Carlo (MC) method has been applied to simulate activation reactions produced in the control rods inserted into the reactor. The activation is mainly produced in the components of stainless steel tubes containing the absorber. The simulation of the reactions permits to obtain a list of the radionuclides generated and its activity (Ródenas et al., 2010d). Obtained activities are introduced as input into another MC model to estimate doses produced by them (Ródenas et al., 2010d, 2010e). The comparison of simulation results with experimental measurements allows the validation of developed models.

The analysis of obtained results showed that the rod handle is the most irradiated part of the control rod. Therefore, the dose out of the pool can be highly reduced inverting the position of the rod into the storage pool placing the handle at a deeper position under water (Ródenas et al., 2010c).

The developed MC models have been also applied to simulate the activation of other materials, such as manganese oxide or components of a stainless steel sample introduced into training or experimental reactors.

These models, once validated, can be applied to other situations and materials where a neutron flux can be found, not only nuclear reactors.

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For instance, activation analysis with an Am-Be source, neutrography techniques in both medical applications and non-destructive analysis of materials, civil engineering applications using a Troxler, analysis of materials in decommissioning of nuclear power plants, etc.

2. Methodology

2.1. Neutron activation

The activity A_j generated in neutron reactions depends on reaction cross sections σ(E), neutron spectrum χ(E), neutron flux distribution Φ(E), concentration of precursors of each radionuclide X_n, irradiation time t_i and a normalization factor C depending on the target concentration. After irradiation, activities decrease with cooling time t_c and disintegration constants, λ_j.

The interaction rate Q_j (reactions/cm^3 s) is given by Eq. (1):

$$Q_j = C\int \Phi(E)\sigma(E)dE \tag{1}$$

where

C is a normalization factor for the atom density (atoms/barn-cm) depending on the target concentration;

Φ(E) is the neutron flux (n/cm^2 s); and

σ(E) is the microscopic cross section of the reaction (barn).

For the isotope j of an element n contained in a target sample, the normalization factor C can be determined from density, composition and isotopic abundances:

$$C = (\rho/M)(X_n/100)(X_j/100)10^{-24}N_A \tag{2}$$

being

ρ density of the sample (g/cm^3).
M atomic or molecular weight (g/mol).
X_n percentage of element n in the sample.
X_j percentage of isotope j in element n.
10^{-24} equivalence for cross section units (cm^2/barn).
N_A Avogadro number (atoms/mol).

Therefore, C is obtained in (atoms/barn-cm).

A matter balance can be done for each j-isotope generated:

$$\frac{dN_j}{dt} = Q_j - \lambda_j N_j \tag{3}$$

and integrating, the concentration N_j (nuclei/cm^3) of the j-isotope is obtained, being t_i the irradiation time:

$$N_j(t) = \left(\frac{Q_j}{\lambda_j}\right)(1-e^{-\lambda_j t_i}) \tag{4}$$

For a cooling time t_c, this concentration N_j becomes:

$$N_j(t) = \left(\frac{Q_j}{\lambda_j}\right)(1-e^{-\lambda_j t_i})e^{-\lambda_j t_c} \tag{5}$$

and the activity can be obtained multiplying by λ_j:

$$A_j(t) = Q_j(1-e^{-\lambda_j t_i})e^{-\lambda_j t_c} \tag{6}$$

$A_j(t)$ is a volumetric activity (Bq/cm^3). To obtain the total activity it is necessary to multiply by the cell volume. The maximum activity will be the asymptotic value, that is, the saturation activity, Q_j, considering an irradiation time very long (~∞) and neglecting the cooling time.

2.2. Monte Carlo models

A Monte Carlo model has been developed using MCNP5 to simulate the activation process (Ródenas et al., 2010b, 2010d). The interaction rate Q_j (Eq. (1)) is calculated using a fluency tally (F4) and an FM4 (tally multiplier card), which provides data for the reactions produced that will be included in the calculation.

The energy spectrum of fission neutrons (Lamarsh and Baratta, 2001) used for the simulation is the Watt distribution described by Eq. (7).

$$\chi(E) = 0.543e^{-1.036E}\sinh(2.29E)^{1/2} \tag{7}$$

The Watt fission spectrum can be considered as a Maxwellian spectrum from a moving reference system (Froehner and Spencer, 1980). The Maxwell fission spectrum alone describes the energy distribution of neutrons emitted by the fission fragments. This does however not include the kinetic energy of the fission ion fragments themselves. As both fission fragments are positively charged, they repel each other due to Coulomb force. This results in kinetic energy of the fission fragments. The Watt spectrum considers the Maxwellian distribution plus the fact that neutrons are emitted from moving fragments. Thus, it is more accurate than the Maxwellian spectrum alone. It is a continuous spectrum with an average energy of 1.98 MeV.

All tallies obtained with MCNP are normalized to be per starting particle. Therefore, activity is calculated per emitted neutron and per second, and it should be multiplied by the instantaneous neutron population that can be calculated as:

$$\dot{N} = \overline{P}\, c\, \nu \tag{8}$$

where

$\dot{N}$ is the instantaneous neutron population (n/s);
$\overline{P}$ is the mean power (W);

C is equal to 3.12*10^{10} fissions/W-s; and

ν is the mean number of neutrons emitted per fission, 2.47 for U-235.

Results for each radionuclide j generated by the neutron activation are the following:

- the interaction rate Q_j (reactions/cm^3-s), Eq. (1), that is, the F4 tally obtained with MCNP5.
- the volumetric activity, A_j (Bq/cm^3).
- activity (per neutron/s emitted at the source) equal to the volumetric activity times the volume of the sample, (Bq).
- total activity (Bq) considering instantaneous neutron population during the irradiation.

All of them can be managed with an Excel sheet.

To estimate the dose rate around irradiated material, another MCNP5 model was developed (Ródenas et al., 2010a, 2010e). Source data for the new input are obtained from the irradiation output, choosing gamma emitters among generated nuclides N_i, with energy E_{ij}, and intensity β_{ij} (branching ratio) of each photopeak j.

Again, the F4 tally was used, now with the FMESH card that allows the user to define a mesh tally superimposed over the problem geometry. Hence, with F4MESH, fluence (cm^{-2}) in nodes of a mesh is obtained. If the source is expressed in photons/s, that is, the activity A_i (dps) times the branching ratio β_{ij} (ph/d), the tally will be obtained in particle flux (cm^{-2} s^{-1}).

Using the DF4 card with appropriate conversion factors, air energy-mass absorption coefficients μ_{en}/ρ extracted from National Institute of Standards and Technology (NIST) for each photon energy of interest and multiplying by this energy, dose rate in MeV/g-s can be obtained (Seltzer, 1993). By means of an appropriated constant for conversion of units, dose rate can be expressed in μSv/h per emitted photon, taking into account that for photons 1 Sv=1 Gy.

3. Applications

3.1. BWR control rods

Neutron activation of control rods in a BWR and doses around the