Joint International Conference on Supercomputing in Nuclear Applications and Monte Carlo 2010 (SNA + MC2010)
Hitotsubashi Memorial Hall, Tokyo, Japan, October 17-21, 2010

Validation of the Monte Carlo Model Developed to Estimate the Neutron Activation of Stainless Steel in a Nuclear Reactor

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Control rods in a nuclear reactor are activated by neutron irradiation. The activity generated will produce a dose around the rod, irrelevant while it is inside the reactor, but significant when the rod is withdrawn from the reactor and placed in a storage pool. This dose is a potential risk for workers in the surrounding area. In previous works, neutron activation and doses have been calculated for a Boiling Water Reactor using the MCNP5 code based on the Monte Carlo method. The model was validated comparing simulation results with experimental measurements in a NPP. On the other hand, most of the activation is produced in stainless steel components of the rod. Therefore, the Monte Carlo model can be also validated considering the activation produced in a piece of stainless steel exposed to some neutron flux in a reactor and measuring the dose around this piece. In this paper, Monte Carlo models developed for both activation of a piece of stainless steel and dose assessment are presented. Comparison of simulation results with experimental measurements in the Training Reactor AKR-2 of the Technical University Dresden shows a good agreement. Hence, the validation of Monte Carlo models can be confirmed.

KEYWORDS: neutron irradiation, Monte Carlo, stainless steel activation, dose assessment, nuclear reactor

I. Introduction

Control rods are activated by neutron reactions into the reactor. The activity so generated will produce a dose around the rod, irrelevant while it is inside the reactor, but it has to be taken into account when the rod is withdrawn from the reactor and placed in a storage pool. This dose is a potential risk for workers in the surrounding area.

Activation reactions can be simulated with the MCNP5 code, [1)] based on the Monte Carlo (MC) method and the number of reactions calculated can be converted into activity. In previous works [2-5)], several MC models were developed to estimate the activity generated as well as the dose around the storage pool. These models were validated by comparing simulation results with experimental measurements in a NPP with Boiling Water Reactor.

On the other hand, most of the activation is produced in stainless steel components of the rod. Therefore, the MC models developed can be also validated considering the activation produced in a piece of stainless steel exposed to some neutron flux in a reactor and measuring the dose around this piece.

In this paper, MC models developed for both the neutron activation of a piece of stainless steel and dose assessment around the irradiated sample are presented. Comparison of simulation results with experimental measurements in the Training Reactor AKR-2 of the Technical University Dresden shows a good agreement. Hence, the validation of MC models can be confirmed.

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II. Methodology

1. Activation

The activity generated in neutron reactions depends on reaction cross sections, neutron spectrum, neutron flux distribution, concentration of precursors of each radionuclide, and irradiation time. After irradiation, activities decrease with time and disintegration constants.

The interaction rate Q (reactions /cm^3-s) is given by:

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

being

- C a normalization factor (at/barn-cm) depending on the target concentration;
- $\Phi(E)$ the neutron flux (n/cm^2-s); and
- $\sigma(E)$ the microscopic cross section of the reaction (barn).

On the other hand, for each j-isotope generated, a matter balance can be done:

$$\frac{dN_j}{dt} = Q_j - \lambda_j N_j \tag{2}$$

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

$$N_j(t) = \left(\frac{Q_j}{\lambda_j}\right)\left(1 - e^{-\lambda_j t_i}\right) \tag{3}$$

For a cooling time t_c the concentration N_j becomes:

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

and multiplying by λ_j to obtain activity:

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

It 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, Q_j, considering an irradiation time very long ($\sim\infty$) and neglecting the cooling time.

2. Experimental Measurements

A cylindrical sample (diameter 20 mm, length 70 mm) has been irradiated during 10 hours into the small zero power reactor AKR of the TU Dresden [6)]. The reactor core is cylindrical (diameter 25 cm, height 27 cm) and has a central horizontal irradiation channel (inner diameter of 24 mm and length equal to core diameter) where the sample has been introduced. A horizontal cross section of the reactor can be seen in **Figure 1**. The stainless steel is X8CrNiTi18.10, whose composition (% in weight) is listed in **Table 1**. [7)]

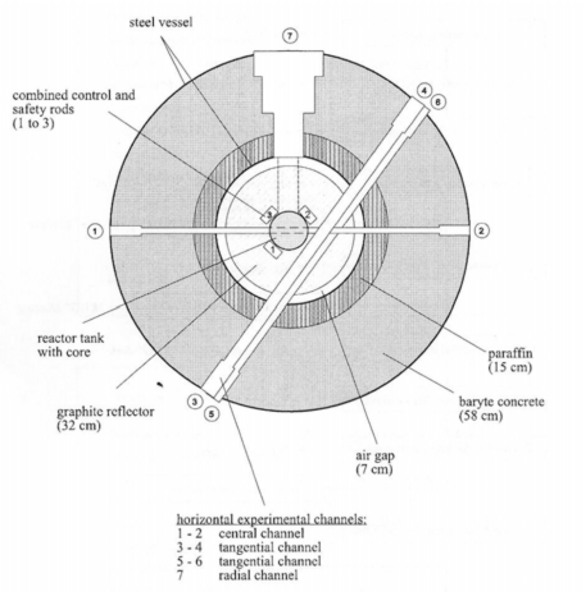

Fig. 1 Horizontal cross section of reactor at core level.

Measured power of the reactor is 0.59 W and maximum thermal neutron flux is 2.5E+07 n/cm^2s. Gamma dose rate in air, in dependence on distance to cylinder has been measured 10 and 30 min after irradiation.

Table 1 Composition of X8CrNiTi18.10.

Element	%
Cr	19
Ni	12
C	0.1
Si	1
Mn	2
P	0.045
S	0.015
Ti	0.4
Fe	65.44

3. Activation Model

The interaction rate Q is calculated by MCNP using F4 tally and FM4 (tally multiplier card), which provides data for the reactions included in the calculation, listed in **Table 2**.

Table 2 Reactions produced in the sample.

P^{31} (n, γ) P^{32}	Fe^{58} (n, γ) Fe^{59}
Cr^{50} (n, γ) Cr^{51}	Ni^{58} (n, γ) Ni^{59}
Cr^{54} (n, γ) Cr^{55}	Ni^{58} (n, α) Fe^{55}
Mn^{55} (n, 2n) Mn^{54}	Ni^{60} (n, p) Co^{60}
Mn^{55} (n, γ) Mn^{56}	Ni^{62} (n, γ) Ni^{63}
Fe^{54} (n,p) Mn^{54}	Ni^{64} (n, γ) Ni^{65}
Fe^{54} (n, γ) Fe^{55}	Mo^{92} (n, γ) Mo^{93}

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

$$\dot{N} = \overline{P}\, c\, \nu \tag{6}$$

where

$\dot{N}$ is instantaneous neutron population (n/s)

$\overline{P}$ is the mean power (W)

c is equal to 3.12E+10 fissions/W-s; and

ν is the mean number of neutrons emitted per fission, equal to 2. 47 neutrons/fission.

For the measured power of 0.59 W, a neutron population equal to 4.55 E+10 n/s is obtained.

4. Dose Rate Assessment

Dose rate at different distances from the irradiated sample have been calculated using again the MCNP5 code and the F4 tally, now with the FMESH card that allows the user to define a mesh tally superimposed over the problem geometry. Hence, with F4MESH, fluence in nodes of a mesh (in cm^{-2}) has been obtained. If the source is expressed in photons/s, the tally will be obtained in particle flux ($cm^{-2}\ s^{-1}$).

Using the DF4 card with appropriated conversion factors, μ_{en}/ρ, extracted from National Institute of Standards and Technology (NIST) [8)] for each photon energy of interest and multiplying by this energy, dose rate in MeV/g-s can be obtained. By means of an appropriated constant for conversion of units, dose rate can be expressed in μSv/h.

As stated above, MCNP results are always normalized to be per starting particle. So they must be multiplied by the number of photons/s emitted by the sample. That number can be obtained from the activity in Bq and the intensity (photons/disintegration) of each photopeak, data extracted from JANIS database. [9)]

The calculated total number of photons/s is 1.66833E+07 for t_c=10 min. On the other hand, the sum of the photons/s emitted by Ni^{65}, Mn^{56} and Cr^{51} together is 1.66826E+07, practically 100% of the total. Furthermore, 98.20% of these photons correspond to the emission of the 3 main lines of Mn^{56}, listed in **Table 3**. Therefore, only Mn^{56} and only these 3 lines will be considered for dose assessment. A similar analysis can be done for t_c=30 min with the same conclusion.

Table 3 Intensities of the main photons emitted by Mn^{56}.

E (MeV)	Intensity
2.11305	0.1433615
1.810719	0.2718925
0.846754	0.9887

III. Results and Discussion

Results from MC simulation are listed in **Tables 4** and **5** respectively for cooling times of 10 and 30 min.

Table 4 Activities calculated for t_c=10 min

nuclide	Tally F4	A (Bq/cm^3)	A (Bq) *per neutron*	A $_{total}$ (Bq)
P^{32}	7.777E-09	1.556E-10	3.422E-09	1.56E+02
Cr^{51}	5.793E-06	6.004E-08	1.320E-06	6.00E+04
Cr^{55}	6.088E-08	8.691E-09	1.911E-07	8.69E+03
Mn^{54}	2.226E-08	2.056E-11	4.522E-10	2.06E+01
Mn^{56}	**1.310E-05**	**1.167E-05**	**2.566E-04**	**1.17E+07**
Fe^{55}	4.274E-06	1.252E-09	2.754E-08	1.25E+03
Fe^{59}	1.061E-07	6.846E-10	1.505E-08	6.84E+02
Co^{60}	3.902E-10	5.855E-14	1.288E-12	5.85E-02
Ni^{59}	1.594E-05	1.577E-13	3.467E-12	1.58E-01
Ni^{63}	2.733E-06	2.162E-11	4.754E-10	2.16E+01
Ni^{65}	6.985E-08	6.246E-08	1.373E-06	6.24E+04
Mo^{93}	2.648E-06	6.983E-13	1.536E-11	6.98E-01

In both tables, the following results are presented for each radionuclide produced in the sample by neutron activation as listed in table 2:

- Tally F4 obtained with MCNP5;
- volumetric activity calculated with equation (5);
- activity (per neutron/s emitted at the source) equal to the volumetric activity times the volume of the sample; and
- total activity (Bq) considering the instantaneous neutron population at the irradiation.

Table 5 Activities calculated for t_c=30 min

nuclide	Tally F4	A (Bq/cm^3)	A (Bq) *per neutron*	A $_{total}$ (Bq)
P^{32}	7.777E-09	1.555E-10	3.420E-09	1.55E+02
Cr^{51}	5.793E-06	6.002E-08	1.320E-06	6.00E+04
Cr^{55}	6.088E-08	1.771E-10	3.895E-09	1.77E+02
Mn^{54}	2.226E-08	2.056E-11	4.522E-10	2.06E+01
Mn^{56}	**1.310E-05**	**1.067E-05**	**2.347E-04**	**1.07E+07**
Fe^{55}	4.274E-06	1.252E-09	2.754E-08	1.25E+03
Fe^{59}	1.061E-07	6.844E-10	1.505E-08	6.84E+02
Co^{60}	3.902E-10	5.855E-14	1.288E-12	5.85E-02
Ni^{59}	1.594E-05	1.577E-13	3.467E-12	1.58E-01
Ni^{63}	2.733E-06	2.162E-11	4.754E-10	2.16E+01
Ni^{65}	6.985E-08	5.699E-08	1.253E-06	5.70E+04
Mo^{93}	2.648E-06	6.983E-13	1.536E-11	6.98E-01

It can be observed in tables 4 and 5 that the most important activity is obtained for Mn^{56}. It is due to the high cross section for the reaction (n, γ) in manganese and to the short half-life of Mn^{56} (2.582 h) that permits this radionuclide to reach equilibrium during an irradiation time of 10 hours.

The longer half-lives of some radionuclides, like Ni^{59} (8E+04 a), Ni^{63} (100 a) and Mo^{93} (3000 a) causes that a low activity is generated because equilibrium is far to be reached.

Co^{60} is a similar case with high half-life (5.272 a), but a low thermal cross section for (n, p) reaction also contributes to strongly decrease the activity produced in the neutron activation.

Really some of the radionuclides produced in the neutron activation can be canceled from the list appearing in tables 4 and 5. Thus, P^{32}, Ni^{59}, Ni^{63} and Mo^{93} can be discarded for dose assessment as they are not gamma emitters. Mn^{54} and Co^{60} can also be neglected a cause of the low activity generated in the irradiation. Cr^{51}, Cr^{55} and Fe^{55} will be also excluded due to the low intensity of the photons emitted. [9)]

Finally, only Mn^{56}, Fe^{59} and Ni^{65} will remain. As stated before (see II.4) they produce practically 100% of all photons emitted by the irradiated sample. Furthermore, the three main lines of Mn^{56} are the most important with 98.2% of all photons emitted by the irradiated sample. These photons are the most rant for dose production and they will be the unique used in dose calculations. This importance is due to the combined effect of higher activity generated in the irradiation and higher intensity of these photopeaks.

A comparison of the activities generated after cooling times of 10 and 30 min is done and the ratio A_{30}/A_{10} is listed in **Table 6**. It can be seen that only Cr^{55} with half-life equal to 3.56 min undergoes an important reduction of activity. Mn^{56}, and Ni^{65} with half-life of some hours experiment decreases of about 10%, while the activity for the rest of radionuclides remains unaltered or with decreases of about 0.1% (when half-life is some days).

Table 6 Comparison of activities for t_c=10 and 30 min

nuclide	A_{30}/A_{10}
P^{32}	0.9993
Cr^{51}	0.9997
Cr^{55}	0.0204
Mn^{54}	1.0000
Mn^{56}	0.9144
Fe^{55}	1.0000
Fe^{59}	0.9998
Co^{60}	1.0000
Ni^{59}	1.0000
Ni^{63}	1.0000
Ni^{65}	0.9124
Mo^{93}	1.0000

Results for dose rates (in μSv/h) at different distances from the irradiated sample are presented in **Tables 7 and 8** for cooling times of 10 and 30 min respectively.

Table 7 Comparison of doses for t_c=10 min.

Distances cm	experimental doses	MCNP doses	ratio MCNP/exp
10	160	170.53	1.07
20	50	46.94	0.94
30	23	20.76	0.90
40	13	11.82	0.91
50	8	7.81	0.98
60	5	5.36	1.07
80	3	2.88	0.96
100	2	2.15	1.07

Experimental measurements and the ratio MCNP/exp are also listed on these tables. A maximum deviation of 9% between simulation and measurement is observed for a cooling time of 10 min, while for 30 min the maximum deviation is 11%.

Therefore, it can be considered as validated the MC models developed.

Table 8 Comparison of doses for t_c=30 min.

Distances cm	experimental doses	MCNP doses	ratio MCNP/exp
10	150	155.94	1.04
20	45	42.92	0.95
30	20	18.98	0.95
40	11	10.81	0.98
50	8	7.14	0.89
60	5	4.90	0.98
80	2.75	2.63	0.96
100	1.8	1.96	1.09

III. Conclusion

A MC model developed to simulate the neutron activation in a reactor has been applied to calculate the activation of a stainless steel sample irradiated in the AKR-2 reactor of TU Dresden.

A MC model has been also used with the MCNP5 code to assess dose rates at different distances from the irradiated sample, some times after irradiation.

Results of the simulation have been compared with experimental measurements and maximum discrepancies for distances up to 1m from the sample are respectively 9% and 10% for cooling times of 10 and 30 min after the irradiation.

These results permit to confirm the validation of MC models developed.

Further developments are foreseen for the next future in order to perform measurements in other experimental or training reactors and compare measurements with MC simulation results.

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