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Abstract: All materials present in the core of a nuclear reactor are activated by neutron irradiation. The activity so generated produces a dose around the material. This dose is a potential risk for workers in the surrounding area when materials are withdrawn from the reactor. Therefore, it is necessary to assess the activity generated and the dose produced. In previous works, neutron activation of control rods and doses around the storage pool where they are placed have been calculated for a Boiling Water Reactor using the MCNP5 code based on the Monte Carlo method. Most of the activation is produced indeed in stainless steel components of the nuclear reactor core not only control rods. In this work, a stainless steel sample is irradiated in the Training Reactor AKR-2 of the Technical University Dresden. Dose measurements around the sample have been performed for different times after the irradiation. Experimental dosimetric values are compared with results of Monte Carlo simulation of the irradiation. Comparison shows a good agreement. Hence, the activation Monte Carlo model can be considered as validated.

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APPLICATION OF DOSIMETRY MEASUREMENTS TO ANALYSE THE NEUTRON ACTIVATION OF A STAINLESS STEEL SAMPLE IN A TRAINING NUCLEAR REACTOR

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1. Introduction

All materials present in the core of a nuclear reactor can be activated by neutron irradiation. When activated materials are withdrawn from the reactor, a dose is produced around them. This dose is a potential risk for workers and people staying in the surrounding area. Therefore, it is necessary to assess the activity generated and the dose produced.

In previous works [1-5], neutron activation of control rods and doses around the storage pool where control rods are placed have been calculated for a Boiling Water Reactor using the MCNP5 code [6] based on the Monte Carlo method.

On the other hand, most of the activation is produced in stainless steel components of the control rod. Indeed, many components in the nuclear reactor core are made of stainless steel. Therefore, the Monte Carlo model can be applied to the activation produced in a piece of stainless steel exposed to some neutron flux in a reactor. As well the dose rate around the activated piece can be measured.

In this work, a stainless steel sample is irradiated in the Training Reactor AKR-2 [7] of the Technical University Dresden. Dose measurements around the sample have been performed for different times after the irradiation.

Experimental dosimetric values are compared with results of the Monte Carlo simulation and the comparison shows a good agreement. It is an attempt to have an indirect validation of the Monte Carlo model for the neutron activation. That is, activities obtained with the Monte Carlo model of the neutron activation are used as input data for a second Monte Carlo model simulating the dose produced around the irradiated piece. These doses are compared with dosimetry measurements. As comparison shows a good agreement between measured and simulated doses, the activation Monte Carlo model can be considered as validated.

2. Methodology

2.1 Neutron 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:

$$Q = C \int \Phi(E)\, \sigma(E)\, dE \qquad (1)$$

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:

$$\frac{dN_j}{dt} = Q_j - \lambda_j N_j \qquad (2)$$

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

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

For a cooling time t_c the concentration N_j becomes:

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

and multiplying by λ_j to obtain activity:

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

$A_j(t)$ 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, Q_j, considering an irradiation time very long (~∞) and neglecting the cooling time.

2.2 Experimental measurements

The training reactor AKR2 [7], acronym for Ausbildungskernreaktor 2, is located at the Technical University in Dresden, Germany. It is a zero power, thermal reactor moderated by solid polyethylene. The fuel elements consist of a homogeneous mixture of moderator and uranium oxide fuel enriched 19.8%. It has a maximum power of 2 W and the maximum neutron flux in the central experimental channel is Φ_{max} = 5 E+07 n/cm^2 s.

The active zone of the core is made up of disk shaped fuel elements with a diameter of 25 cm. The height of the active zone is 27.5 cm.

For the experiment, the reactor is driven at a power level of P=0.59 W. This corresponds to a measured neutron flux of 2.5 E+07 n/cm^2 s. This flux has been measured in the central experimental channel of the reactor. The cross section of the whole reactor is shown in figure 1.

A stainless steel sample type X8CrNiTi18.10 is irradiated for 10 hours in the central experimental channel of the reactor. The sample has cylindrical shape with a radius of 1 cm and a length of 7 cm. It has a volume of 21.99 cm^3 and a density ρ= 7.9 g/cm^3. Thus it has a mass of 173.73 g. The composition of the sample is listed in Table 1 [8].

Dose experimental measurements were done with a Berthold dose rate meter type LB133-1 equipped with an ionisation chamber detector LB6006, suitable for photon dose equivalent measurements in photon energy range 30 keV - 1.3 MeV. The device was calibrated by official authorities.

2.3 Monte Carlo model

An activation Monte Carlo model has been developed using MCNP5. The interaction rate Q (eq. 1) is calculated using F4 tally and FM4 (tally multiplier card), which provides data for the reactions included in the calculation, listed in Table 2.

The energy spectrum of fission neutrons [9] used for the simulation is the Watt distribution described by equation 6.

$$\chi(E) = 0.453\ e^{-1.036E}\ \sinh\left(2.29\ E\right)^{1/2} \qquad (6)$$

The Watt fission spectrum can be considered as a Maxwellian spectrum from a moving reference system [10]. 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 MCNP5 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

$$\dot{N} = \overline{P}\, c\, \nu \qquad (7)$$

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.

2.4 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 (cm^{-2}) in nodes of a mesh 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, air energy-mass absorption coefficients μ_{en}/ρ extracted from National Institute of Standards and Technology (NIST) [11] 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 per emitted photon, taking into account that for photons 1 Sv = 1 Gy.

As stated above, MCNP5 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 [12].

It has been simulated 100 million particles to obtain relative errors lower than 0.3 %. MODE P, E has been activated to follow tracks of photons and electrons. A cutoff of 5 keV for electrons has been used in order to reduce the computation time.

3. Results and discussion

3.1 Activity

Results from MCNP5 simulation are listed in Tables 3 and 4 respectively for cooling times of 10 and 30 min. 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 calculated with equation (7).

It can be observed in Tables 3 and 4 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} (80,000 y), Ni^{63} (100 y) and Mo^{93} (3,000 y) 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 y), Furthermore, cobalt is not present in X8CrNiTi18 stainless steel being Co^{60} only produced by the (n, p) reaction in Ni^{60} with a low thermal cross section that contributes to strongly decrease the activity produced in the neutron activation.

Really many of the radionuclides produced in the neutron activation can be canceled from the list appearing in Tables 3 and 4. Thus, P^{32}, Ni^{59}, Ni^{63} and Mo^{93} can be discarded for dose assessment as they are not gamma emitters. Mn^{54}, Fe^{59} and Co^{60} can also be neglected a cause of the low activity generated in the irradiation. Cr^{55} and Fe^{55}will be also excluded due to the low intensity of the photons emitted [12].

Finally, only Cr^{51}, Mn^{56}, and Ni^{65} will remain. They produce practically 100% of all photons emitted by the irradiated sample. Furthermore, 98.20% of these photons correspond to the emission of the 3 main lines (2.11305; 1.810719; and 0.846754 MeV) of Mn^{56}. Therefore, only Mn^{56} and only these 3 lines will be considered for dose assessment. Its relevance is due to the combined effect of higher activity generated in the irradiation and higher intensity of emitted photons corresponding to these photopeaks.

3.2 Dose

Doses calculated by MCNP5 and measured doses are compared in Tables 5 and 6 respectively for cooling times of 10 and 30 minutes. A good agreement between the experimental and simulated values can be seen. For 10 minutes cooling time, the maximal deviation between MCNP5 and experiment is 8%. For a cooling time of 30 minutes the maximal deviation is 12%.

The MCNP5 values are partly above and partly below the measured values. Thus there is not a systematic error as for example an offset deviation. In average MCNP5 results tend to be below measured doses.

4. Conclusions

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

Results of the simulation have been compared with experimental measurements.

Maximum discrepancies for distances up to 1 m from the sample are respectively 8 % and 12 % for cooling times of 10 and 30 min after the irradiation.

These results permit to confirm the validation of Monte Carlo 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 Monte Carlo simulation results.

Irradiation of reactor components in power reactors is very long compared to the time of irradiation of the sample in AKR2. A component in a reactor is irradiated up to 15 years in average. Thus, also radionuclides with high half life build up during the irradiation in power reactors, but they hardly appear in the current problem. However, this is not downgrading the model.

As it works for short lived nuclides, it will also be applicable for long lived nuclides.

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Table 1. Composition of the sample.

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Element	Weight fraction %	Molar mass g/mol
Cr	19.000	51.9961
Ni	12.000	58.6934
C	0.100	12.0107
Si	1.000	28.0855
Mn	2.000	54.938
P	0.045	30.973
S	0.015	32.065
Ti	0.400	47.867
Fe	65.440	55.845

Table 2. Reactions produced in the sample.

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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}

Table 3. Activities calculated for tc=10 min.

Table 3. 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

Table 4. Activities calculated for tc=30 min.

Table 4. Activities calculated for t_c=30 min

			A (Bq)	
nuclide	**Tally F4**	**A (Bq/cm^3)**	***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

Table 5. Doses after t_{cool}=10 min

Distance (cm)	MCNP (µSv/h)	Measured (µSv/h)	Ratio MCNP/measured
10	162	160	1.01
20	45.8	50	0.92
30	20.9	23	0.91
40	11.9	13	0.92
50	7.8	8	0.98
60	5.4	5	1.08
80	3.1	3	1.03
100	2	2	1

Table 6. Doses after tcool=30 min.

Table 6. Doses after t_{cool}=30 min

Distance (cm)	MCNP (μSv/h)	Measured (μSv/h)	Ratio MCNP/measured
10	148.4	150	0.99
20	41.9	45	0.93
30	19.2	20	0.96
40	10.9	11	0.99
50	7.12	8	0.89
60	4.9	5	0.98
80	2.8	2.5	1.12
100	1.85	1.8	1.03

Figure 1. Cross section of the whole reactor.
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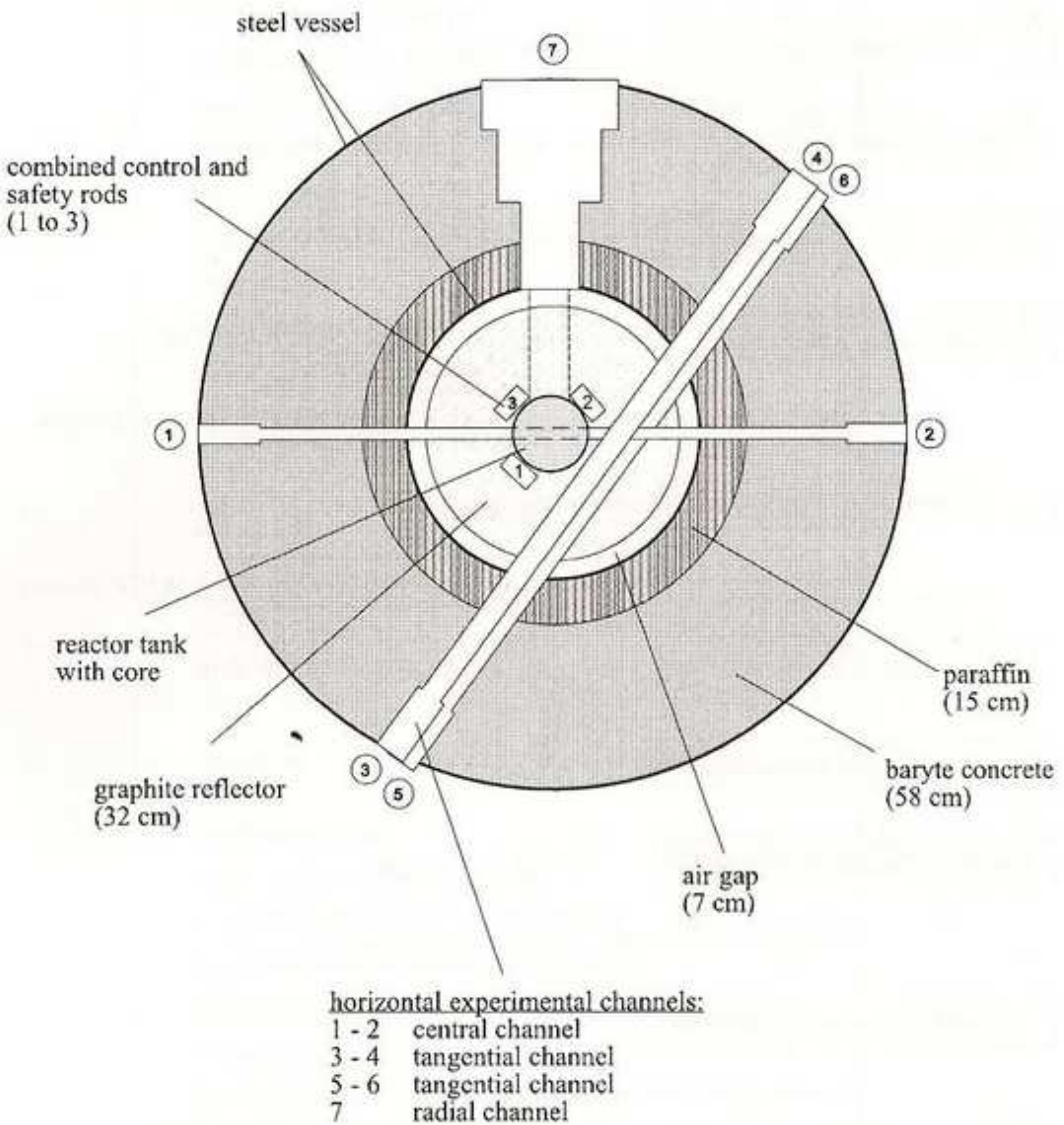

*Highlights (for review)

All materials present in the core of a nuclear reactor can be activated by neutron irradiation.

Neutron activation can be simulated using the MCNP5 code based on the Monte Carlo method.

Most of the activation is produced in stainless steel components of the nuclear reactor core.

A stainless steel sample is irradiated in a reactor and doses are measured around the sample.

Experimental dosimetric values are compared with results of the Monte Carlo simulation.