Proc. of the Int. Workshop on Nuclear Data for the Transmutation of Nuclear Waste, GSI-Darmstadt, Germany, Sept. 1-5, 2003, ISBN3-00-012276-1.

Testing of Different Data Libraries in Activation Analysis of Concrete and Graphite from Nuclear Installations

M. Cometto, D. Ancius, and D. Ridikas*

CEA Saclay, DSM/DAPNIA/SPhN, F-91191 Gif-sur-Yvette, France

With the aging of the nuclear park, decommissioning and dismantling of nuclear installations after their service life is becoming an important issue for the nuclear industry. The radiological characterisation of the equipment and structures present in the reactor and its environment is an essential stage in a decommissioning project since it permits to define and optimize the decommissioning strategy and the disassembling operations. In addition, correct activation estimates are essential for determining the quantity and the nature of the radiological waste generated during decommissioning. The adoption of efficient dismantling procedures and the optimization of the mass flow going to different waste repositories might reduce substantially the total cost of decommissioning.

The present work has been done in the framework of the decommissioning and dismantling of the experimental reactor of the University of Strasburg (RUS). A methodology that combines theoretical calculations and direct measurements has been developed for determining the long-term induced activity in the graphite, concrete and materials present in the reactor. After characterisation of the different elements present in the reactor, it is then possible to plan efficiently the disassembling and dismantling of the system and to optimise the mass flow going to different waste repositories. From a scientific perspective, the comparison of theoretical predictions with experimental values validates the approach and the methodology used in the present study and tests the consistency and the reliability of the nuclear data employed for activation analysis.

Introduction

Decommissioning of nuclear installations after their service life and their dismantling are connected with the necessity of the disassembling, handling and disposing of a large amount of radioactive equipment and structures. In particular, the concrete used as a biological reactor shield and graphite as a moderator-reflector represent the majority of waste requiring geological disposal. To reduce this undesirable volume to the minimum and to successfully plan the dismantling and disposal of radioactive materials in the storage facilities, the activation of the structures should be accurately evaluated.

In the framework of the decommissioning and the dismantling of the experimental nuclear reactor RUS in Strasbourg (France) detailed activation estimates have been conducted for characterising the graphite and the structural materials present in the reactor environment. For this purpose, the isotopic composition of fresh graphite samples and different types of concrete has been determined by activation analysis in the research reactors OSIRIS and ORPHEE of CEA Saclay (France). The neutron flux distribution has been computed in the whole reactor using Monte-Carlo methods in conjunction with variance reduction techniques. Finally, the spatial activation distributions of graphite, concrete and other materials have been estimated in different reactor zones as a function of the three main nuclear data libraries, i.e. ENDF, JEF and JENDL. In parallel, the activation of representative graphite and concrete samples from RUS has been measured experimentally.

The present paper reports on the preliminary results of this work with particular attention on the possible sources of uncertainty in activation predictions. In particular, we quantify the impact of using different nuclear data libraries in activation calculations, as well as the uncertainty given by the impurity level in graphite and concrete. The comparison of theoretical predictions with experimental values will validate the approach and the methodology used in the present study. In addition, it will test the consistency and the reliability of the nuclear data used for activation analysis.

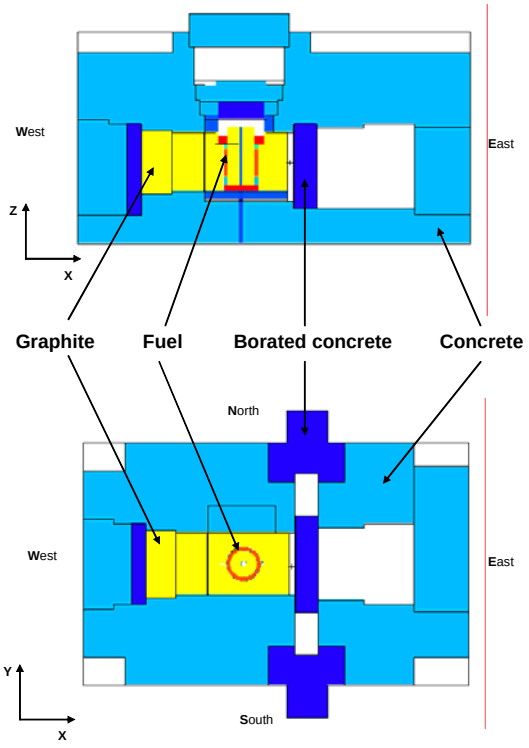
* ridikas@cea.fr

Generalities about the RUS reactor

The University of Strasbourg has built in 1966 the experimental nuclear facility RUS (Reactor of the University of Strasbourg). The reactor had both teaching and irradiation purposes and has been mainly used for creating short-lived radioactive isotopes. The concept of the experimental plant is based on the ARGONAUT design that was developed at Argonne National Laboratory (USA).

RUS is a thermal-spectrum, water-cooled and graphite-moderated reactor with a maximal thermal power of 100 kW. A simplified three-dimensional model of the reactor is presented in Figure 1. The fuel elements consist of aluminium and highly-enriched uranium plates (93% of ^{235}U) and are disposed annularly around an inner graphite reflector. Finally, they are surrounded by an outer graphite reflector. Two additional graphite columns are placed in the west side of the reactor with the purpose of thermalising further the neutron flux. Two neutron flux detectors are inserted in the middle of the inner reflector and at the interface between the fuel region and the outer reflector. Several blocks of concrete, borated or ordinary, have structural purposes and ensure the biological shielding of the core.

Fig. 1 : 3D geometry model of RUS.

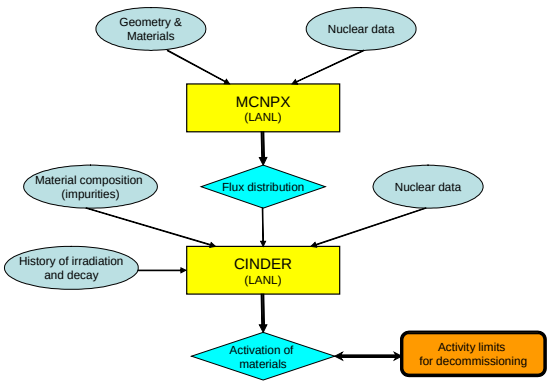


The reactor had the first irradiation at zero-power in November 1966 and started the operations at full power in May 1967. A first set of fuel elements was discharged in 1978 and was replaced by fresh fuel. RUS was definitively shut-down in December 1997 and the fuel elements including control rods have been removed from the core in December 2000. During 31 years of operation, the reactor had an utilisation factor of 14% with an average power of 73 kW. No incidents have been reported during normal operations. The dismantling of the nuclear installation is planned for 2006.

Calculation methods and nuclear data

The calculation of activation in the reactor materials is performed in two separate steps (in yellow), as illustrated in Figure 2. In the first step, spatial neutron flux distributions are determined in the whole reactor for a nominal power of 100 kW. In the second step the "static" neutron flux is combined with the history of irradiation and decay in order to obtain the activation of materials in the whole reactor.

Fig. 2: Scheme for neutron transport and activation calculations.



Neutronic calculation are performed with the Monte-Carlo high-energy transport code MCNP[1)] and using ENDF-B/VI basic nuclear data library. Variance reduction techniques are used to improve the accuracy of neutronic flux estimates in the shielding structures. As shown in Figure 1, the reactor is accurately described using a complete

three-dimensional geometry model. The continuous spatial neutron flux distributions resulting from a MCNP calculation are then condensed in a 63 energy-group structure to be used by the CINDER '90 activation code.

The activation of materials is computed with the deterministic code CINDER '90[2]. CINDER '90 uses its own nuclear data library originating from different sources, mainly from ENDF, JEF and JENDL but also from theoretical models. Essential inputs for the code are the exact isotopic composition of the reactor materials and the history of irradiation and decay. For the latter, the annual averaged values of the neutron flux are used in the simulations. As a result of the activation calculation, the total activity and its isotopic composition are determined at appropriate time steps after the final shut-down of the reactor.

Neutron flux calculations

Neutron flux calculations are performed for a fresh core supposing that the control roads are fully inserted. Figure 3 reports the neutron flux distributions in the XY plane at the middle of the fuel element which corresponds to the maximal axial flux. Both the total and fast (above 1 eV) flux components are presented.

The maximal neutron flux, of $2.8 \cdot 10^{12}\, n/(s \cdot cm^2)$, is located in the annular fuel region, close to the internal reflector. High-energy neutrons from the fuel region are diffused and thermalised by the graphite reflector and are then absorbed in the concrete walls. The large absorption in the boron and the consequent large flux drop in the east and west borated concrete walls are also clearly visible in the same figure. About 10 orders of magnitude in the flux level are effectively represented, up to a value of $10^2 \div 10^3\ n/(s \cdot cm^2)$ in the external shielding.

Neutron spectra in four graphite reflector regions are shown in Figure 4. The internal reflector features the hardest spectrum, with only 54% of thermal neutrons (E < 1 eV). The neutron spectrum becomes softer with the increasing distance from the fuel: the neutron thermal fraction increases from 67% in the outer reflector to 96.5% and 99.9% in the first and second column respectively.

Calculated neutron flux values are in good agreement with the experimental measures in the two detectors, especially for the thermal component. Calculations overestimate the thermal flux by 10% ÷ 15% with respect to measures, while somewhat larger discrepancies are observed for the epithermal and fast components.

Fig. 3: Neutron flux distributions in the XY plane.

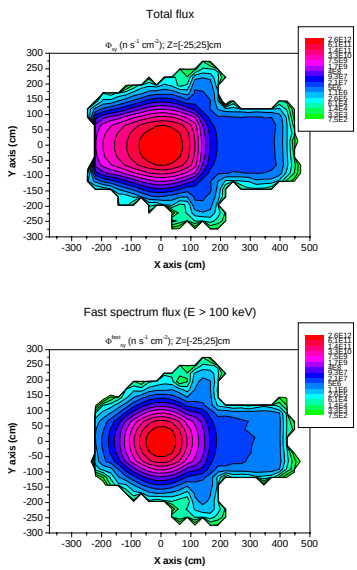
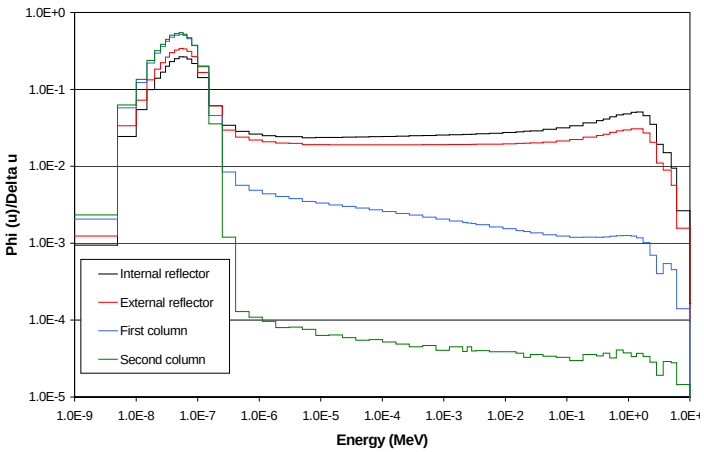


Fig. 4: Neutron spectra in different graphite regions.



Activation calculations

The long-term activation of structures in a nuclear reactor is mainly due to the interactions between thermal neutrons and impurities in the reactor materials. For this reason, the knowledge of the isotopic composition of all materials present in the system is crucial for activation estimates. For a preliminary analysis, the isotopic composition of graphite and different types of concrete is taken from bibliographical references. The exact composition of materials, determined by activation analysis of fresh reactor material sample, will be used in the final phase of the study. The total activity of the inner graphite reflector, including the decomposition by isotopes, is given in Figure 5 as a function of time of dismantling. Figure 6 gives equivalent data for the borated concrete wall at the east of the reflector.

At the beginning, the activity of the graphite reflector is dominated by tritium, which is originated from a (n,α) reaction on ^{6}Li. In a longer run, the principal radioelement is ^{14}C generated by a capture (n,γ) reaction on ^{13}C and by a (n,p) reaction on ^{14}N. The latter becomes dominant for a nitrogen concentration greater than 10 ppm (part per million). Other important contributors are ^{55}Fe, ^{60}Co, ^{152}Eu and ^{154}Eu which are all created by a (n,γ) reaction. With the exception of ^{13}C, all stable isotopes are present in the graphite as impurities, with a concentration varying between 0.1 and 50 ppm.

In the case of the activation on concrete, the main contributors are ^{55}Fe, ^{60}Co, ^{133}Ba, ^{152}Eu and ^{154}Eu, all generated by (n,γ) reactions on stable isotopes. With the exception of iron, all other isotopes are present in the concrete as impurities.

Fig. 5: Activation of inner graphite reflector.

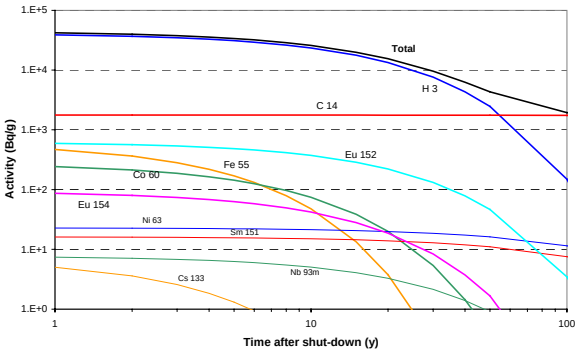
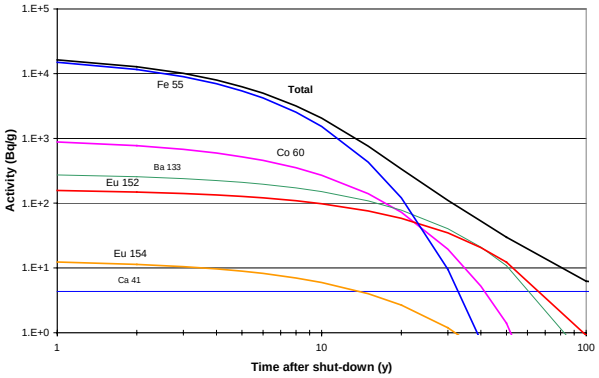
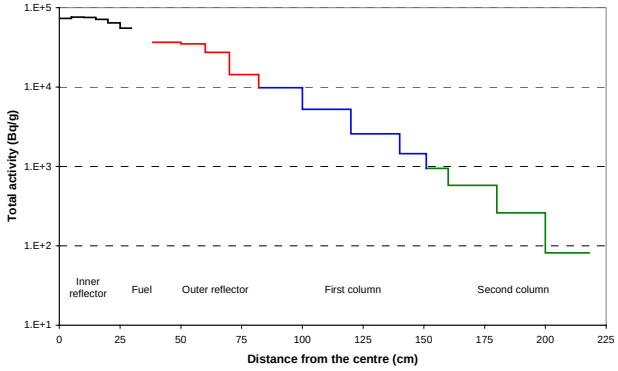


Fig. 6: Activation of heavy borated concrete.



The activity level in a given material is dependent on the fluence and neutron spectrum and therefore reflects the position in the reactor. Figure 7 shows the total activity in the graphite reflector as a function of the distance from the core centre. The activity range in the graphite covers more than three decades from about 10^{5} Bq/g to about 30 Bq/g in the lower part of the second graphite column. As expected, the total activity in the outer reflector and in the two thermal columns decreases exponentially with the distance from the fuel region. A similar trend is not observed in the inner reflector and in the first 20 cm of the outer reflector, due to the strong variations of the neutron spectrum. The higher total flux at the interface with the fuel is compensated by a softer neutron spectrum in the internal graphite regions.

Fig. 7: Spatial activity distribution in the graphite zones.



The total activation decreases exponentially in the concrete walls since the neutron flux is already thermalised before entering concrete.

Uncertainties in activation predictions

The total uncertainty on activation prediction accounts for all the approximations made at each stage of the calculational procedure and therefore depends on:

i) The uncertainty on the neutron flux estimation
ii) The uncertainty on the history of irradiation and decay
iii) The uncertainty on the isotopic composition of reactor materials
iv) The uncertainty on the nuclear data for activation

The main objective of the present section is to quantify the uncertainties in the production rates of radioactive isotopes due to different nuclear data libraries used in activation calculations. For this purpose, ENDF, JEF and JENDL-based activation cross-sections are condensed in a 63 energy-group structure using the continuous neutron flux calculated by MCNP in the inner graphite reflector. The production rates obtained with the three main data libraries are then compared with those of CINDER'90 for the same neutron flux. Table 1 reports the differences on JEF, JENDL and CINDER'90 one-group cross-sections relative to the reference ENDF-B/VI data. Differences of more than 30% are given in red and those of more than 5% in blue.

Table 1: Cross-section differences relative to the ENDF library

Reaction	*JEF*	*JENDL*	*CINDER '90*
Li 6 (n, α) → Tritium	-0.16 %	0.10 %	-1.06 %
C 13 (n, γ) → C 14	-	-	-37.49 %
N 14 (n, p) → C 14	-0.61 %	-3.07 %	94.55 %
Cl 35 (n, γ) → Cl 36	-	Ref. *	-0.56 %
Ca 40 (n, γ) → Ca 41	-	-	-0.42 %
Fe 54 (n, γ) → Fe 55	14.82 %	-3.46 %	-0.36 %
Co 59 (n, γ) → Co 60	-0.20 %	0.05 %	-54.06 %
Ni 62 (n, γ) → Ni 63	-0.18 %	-0.64 %	0.09 %
Cs 133 (n, γ) → Cs 134	-6.67 %	-0.98 %	2.17 %
Eu 151 (n, γ) → Eu 152	-0.19 %	0.06 %	-31.03 %
Eu 153 (n, γ) → Eu 154	-7.07 %	3.33 %	-0.48 %

"*" - Reference library; "–" - no ENDF data available in MCNP format.

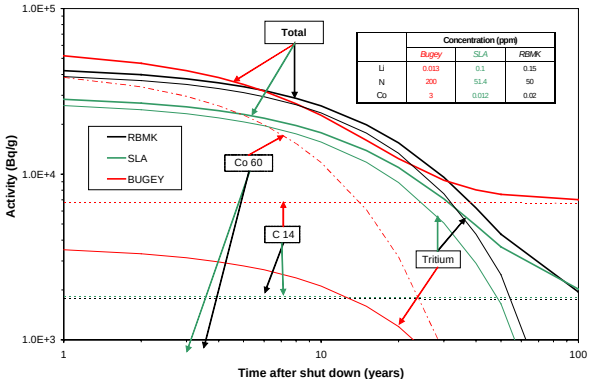
Concerning the most significant reactions for activation estimates, a good agreement is found among the three main nuclear data libraries. In most of the cases, the uncertainty on the production rates is lower than 3%. Larger discrepancies are observed for capture reactions of ^{54}Fe and ^{153}Eu, which lead to a maximal difference of 10-20% on specific activity predictions. On the contrary, the one-group cross sections obtained from CINDER '90 data differ considerably from those based on the other sources, leading to a large underestimation on the ^{60}Co and ^{152}Eu activities and to a great uncertainty on that of ^{14}C. Re-evaluation and update of the CINDER '90 library is therefore necessary.

As already mentioned, the knowledge of the isotopic material composition is essential for accurate activation predictions. Concerning the activation of graphite, a sensitivity study is conducted for three different isotopic compositions that were found in the literature. Two set of data, named SLA and BUGEY thereafter, correspond to graphite used in the nuclear power plants of Saint-Laurent des Eaux and Bugey that were built in the same period as RUS (all three in France). The graphite composition is most likely to be similar to that of RUS, but the bibliographical data available are incomplete and quite old. A third set of data, named RBMK, corresponds to the graphite used in the nuclear power plant of Ignalina (Lithuania). The isotopic composition was recently determined by gamma spectroscopy

analysis and by diffusion mass spectroscopy at CEA Saclay. In addition, measured data are also integrated by bibliographical references. The results of this parametric study are shown in Figure 8.

Large differences are observed in the impurity level of Co, N, Eu and Li which are responsible for more than 90% of the total activity of graphite. As a function of the graphite composition used, the ^{60}Co activity vary by a factor of 300, that of tritium by a factor of 10 and that of ^{14}C by about 4 times. However, due to compensating effects, the uncertainty on the total activity level is within a factor of 2 in the first period after the shut-down. The evolution of the total activity varies considerably with time since it is dominated by different radioelement in the 3 reference cases studied. In all cases the long-term activity is dominated by ^{14}C.

Fig. 8: Activation in the inner reflector for different graphite isotopic compositions.



Conclusions

In the framework of the decommissioning and dismantling of the experimental reactor of the University of Strasburg a preliminary study has been conducted to determine the residual activity on the graphite and other structural materials. The final goal of the present study is to characterise the different elements present in the reactor and to optimise the mass flow going to the waste repositories. The following conclusions can be drawn:

- Neutron flux estimates are in good agreement with the experimental data available near the fuel region and seem to be sufficient for the precision required for decommissioning and dismantling.
- The three main nuclear data libraries, i.e. ENDF, JEF and JENDL, produce consistent results in the activation calculations for the most important radio-elements. On the other hand, large discrepancies are observed when the CINDER'90 data are used, suggesting the need for a library update or a re-evaluation of some specific reactions.
- The precise isotopic composition of the reactor materials can not be inferred from bibliographical references since it depends on the quality of the material used and on the fabrication processes. Experimental measurements of fresh samples of reactor materials are therefore essential for determining the impurity level and reducing the uncertainties on activation estimates.

The determination of the exact isotopic composition of all reactor materials is currently on the way and will allow for more accurate predictions in a second step of this study. In addition, the comparison of numerical predictions with on-site measures of irradiated samples will validate the approach and methodology used in the present study. At the same time it will test the reliability of nuclear data for activation.

According to preliminary results, a large amount of graphite and concrete could be saved or send to very low activity waste repository.

References

[1] J. F. Briesmeister for Group X-6 "MCNP-A, A General Monte Carlo Code for Neutron and Photon Transport", Version 4A, LANL, report LA-12625-M (1993).

[2] W. B. Wilson, T. R. England and K. A. Van Riper "Status of CINDER'90 Codes and Data", Los Alamos National Laboratory, report LA-UR-99-361 (1999).