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Activation analysis of graphite and concrete in the experimental reactor RUS

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Abstract

Decommissioning and dismantling of nuclear installations after their service life 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 to storage facilities, the activation of the structures should be accurately evaluated.

In the framework of the decommissioning and the dismantling of the experimental reactor of the University of Strasbourg, detailed activation estimates have been conducted to characterise the graphite and the structural materials present in the reactor environment. For this purpose, the chemical 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). Then, the activation of graphite, concrete and other materials has been calculated in the whole reactor 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 has been measured experimentally.

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 used for activation analysis. We believe that a similar approach could also be used for the decommissioning of industrial nuclear reactors.

INTRODUCTION

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 materials present in the reactor and its environment is a fundamental stage in a decommissioning project since it permits to define and optimise 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 and storage.

The present work has been done in the framework of the decommissioning and dismantling of the experimental Reactor of the University of Strasbourg (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 metals present in the reactor. The final objective of the study is to characterise the different elements present in the reactor, to optimise the mass flows going to the waste repositories and hence minimise the total cost of dismantling and decommissioning.

GENERALITIES ABOUT THE REACTOR RUS

In 1966 the University of Strasbourg has built the experimental nuclear facility RUS, based on the ARGONAUT reactor core design. RUS is a thermal-spectrum, water-cooled and graphite-moderated reactor with a maximal thermal power of 100 kW. The fuel, constituted by aluminium and highly-enriched uranium (93% of ^{235}U), is mounted annularly around an inner graphite reflector and is 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. The biological shielding is ensured by borated and "heavy" concrete walls. A simplified three-dimensional model of the reactor is presented in Figure 1.

RUS had the first irradiation at full power in April 1967 and was definitively shut-down in December 1997. On average, the experimental facility had an utilisation factor of 14%, with an average power of 73 kW. During 31 years of operation, RUS produced about 2.81 GWh. No accidents have been reported during the life of the installation.

CALCULATIONAL TOOLS AND NUCLEAR DATA

The methodology used for determining the long-term induced activity in the graphite, concrete and other structural materials combines theoretical calculations and direct measurements. As first, the chemical composition of appropriate concrete and graphite samples is measured by activation analysis in the research reactors of OSIRIS and ORPHEE (CEA Saclay). Then, the activation of graphite and structural materials is calculated in the whole reactor building and its surroundings. Finally, the activation of representative graphite and concrete samples is measured experimentally and is compared with the theoretical predictions.

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 the reactor power, irradiation and decay in order to obtain the activation of materials in the whole reactor. As a result of the activation calculation, the total activity and its isotopic decomposition are determined at appropriate time steps after the final shut-down of the reactor.

Neutronic calculations are performed with the Monte-Carlo transport code MCNP [1] and using the ENDF-B/VI basic nuclear-data library. 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 [2].

CINDER '90 uses its own nuclear-data library originated from different sources, mainly from ENDF, JEF and JENDL but also from theoretical models. Due to the large discrepancies observed for

^{13}C, ^{14}N, ^{59}Co and ^{151}Eu cross-sections, the original CINDER 90 data have been replaced with the most recent evaluations from ENDF and JENDL [3]. 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 fluence are used in the simulations.

CHEMICAL COMPOSITION OF REACTOR MATERIALS

Two different samples have been analysed for determining the impurity level in the moderating graphite. An irradiated sample has been drawn from the second column and a fresh graphite brick has been taken from the reserves. It was thought that a fresh sample from reserves could be representative of the reactor materials. As a matter of fact, large differences are observed for the impurity level of several elements, including Fe, Co, Ni and Eu, indicating that the two samples do not have the same origin. The chemical composition of the sample drawn from the graphite column has been used as a reference in the present study. The impurity level of light elements (Li, N, O) has not been measured in the graphite samples. Conservative values from literature are assumed for the activation estimates.

Several samples of concrete have been analysed, either active samples from the reactor or inactive samples from reserves, when the material is not easily accessible. In particular, only two borated concrete samples are drawn directly from the reactor while three additional concrete samples are taken from reserves. The chemical composition of the samples is then compared with the "expected" composition resulting from the specifications followed for concrete manufacture. The chemical composition of the ordinary and "heavy" concrete are coherent with the specifications. On the contrary, large discrepancies are observed for the different borated concretes that surround the core. In particular, the measured concentration of boron and hydrogen differs from the expected value (about a factor of 2) and the barium is present in all the samples as an impurity, while it is expected to constitute a large fraction of some neutron shields. To reduce those uncertainties, additional samples of borated concrete have been recently drawn and are currently analysed.

The differences observed in the composition of graphite samples have an impact on the activation calculations but do not change the neutron flux distributions in graphite and concrete. On the contrary, the uncertainties on hydrogen and boron concentration in the shields may considerably affect the neutron-flux estimations in the concrete.

NEUTRON FLUX DISTRIBUTIONS

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 100 keV) flux components are presented. The maximal neutron flux, of 2.8 10^{12} $n/s \cdot cm^2$ (~50 % thermal), 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 $10^2 \div 10^3$ in the external shielding.

During the reactor operation the neutron flux has been measured in two detectors, inserted in the middle of the inner reflector and at the interface between the fuel region and the outer reflector. Calculated neutron-flux values are in good agreement with the experimental measures, especially for the thermal component, as shown in Table 1. Calculations overestimate the thermal flux by about 15% with respect to measures, while somewhat larger discrepancies are observed for the epithermal and fast components.

ACTIVATION ANALYSIS IN GRAPHITE

The total activity of the first graphite column, including the decomposition by isotopes, is given in Figure 4 as a function of the time of dismantling. The long-term activation of graphite reflector is dominated by tritium, which is originated mainly from a (n,α) reaction on ^{6}Li, and ^{14}C, generated by a

capture (n,γ) reaction on ^{13}C and by a (n,p) reaction on ^{14}N. From a radioprotection viewpoint, other important contributors are ^{60}Co, ^{94}Nb, ^{134}Cs, ^{152}Eu and ^{154}Eu, all generated by capture reactions. With the exception of ^{13}C, all stable isotopes are present in the graphite as impurities, with a concentration varying between 0.1 and 200 ppm. The activity level in a given material is dependent on the fluence and neutron spectrum and therefore reflects the material position in the reactor. The activity range in the graphite covers more than three decades from about $9 \cdot 10^4$ Bq/g to about 10 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.

COMPARISON WITH EXPERIMENTS

In order to validate the numerical estimations, a graphite sample of about 10 cm long has been drawn from the second column and its isotopic activity has been measured. The experimental results are reported in Table 2, together with the calculated values. Only the activity of tritium, ^{14}C and ^{60}Co has been quantified experimentally, while that of all other isotopes is inferior to the respective detection limits, as indicated in the table. The agreement between calculations and experiments is excellent for ^{60}Co, the measured activity being only 15% larger than the calculated. Larger discrepancies (about a factor 2÷3) are observed for tritium and ^{14}C activity predictions. However, those isotopes are produced by reactions on light elements that could not been measured from fresh graphite sample. The discrepancies observed can be explained by the assumptions made on the impurity assumed for Li and N. Concerning all the other isotopes, the calculated activities are coherent (well below) the respective detection limits.

CONCLUSION

The methodology that combines detailed theoretical calculations with representative direct measurements has been developed for determining the long-term activity in the graphite and structural materials present in the experimental reactor RUS of the Strasbourg University. The proposed approach relies on precise knowledge of the nuclear installation, including the detailed plans of the plant, the chemical composition of the materials and the history of irradiation.

The appropriate sampling of reactor materials is a key issue, due to the difficulties on accessing remote part of the plant and managing very active samples. In addition, our experience showed that non-irradiated samples from reserves may not be representative of all "realistic" reactor materials.

The calculated activation in graphite is in excellent agreement with the measurements, indicating that the methodology used can be successfully applied to the whole reactor. However, a better knowledge of the hydrogen and boron concentration in the borated shields is necessary to reduce the uncertainties in the concrete activation.

Finally, the present study showed that a non negligible volume of the two graphite columns (about 4 tonnes) could be sent to a 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, preprint LA-UR-99-361 (1999).

3. M. Cometto, D. Ancius, D. Ridikas, *Testing different data libraries in activation analysis of concrete and graphite from nuclear installations*, Proceedings of the International Workshop on Nuclear Data for the Transmutation of Nuclear Waste, GSI-Darmstadt (Germany), 1-5 September 2003; ISBN3-00-012276-1 (http://www-wnt.gsi.de/TRAMU).

Figure 1: Three-dimensional model of the reactor RUS.

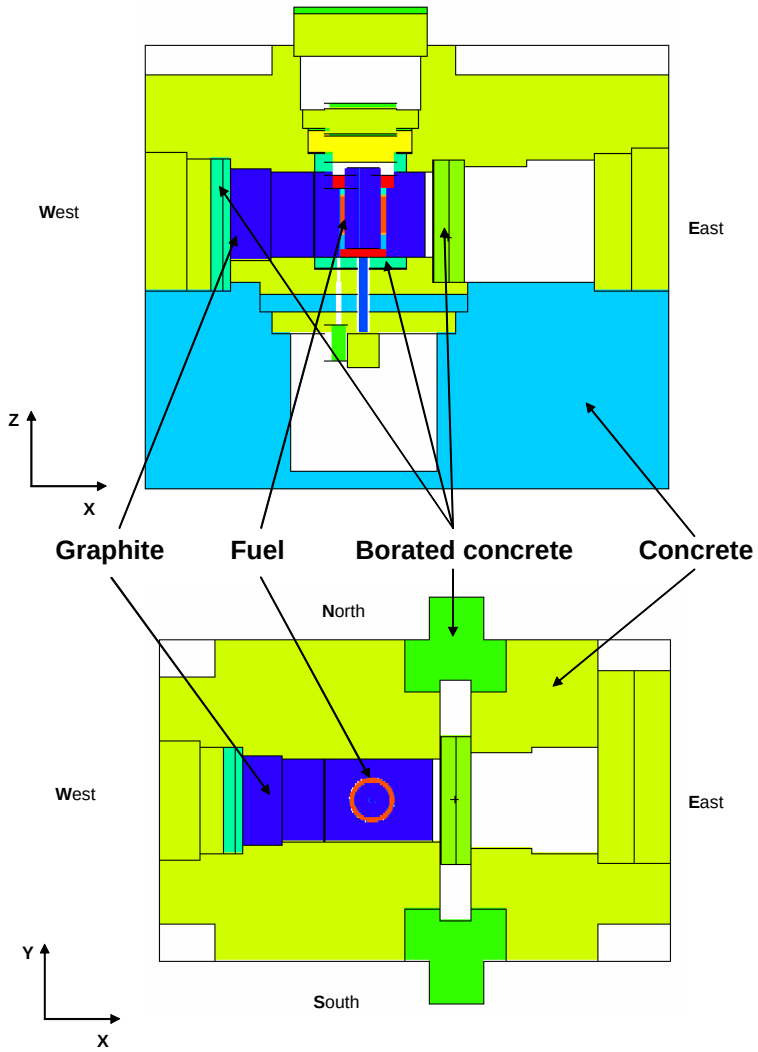

Figure 2: Scheme for neutron transport and activation calculations.

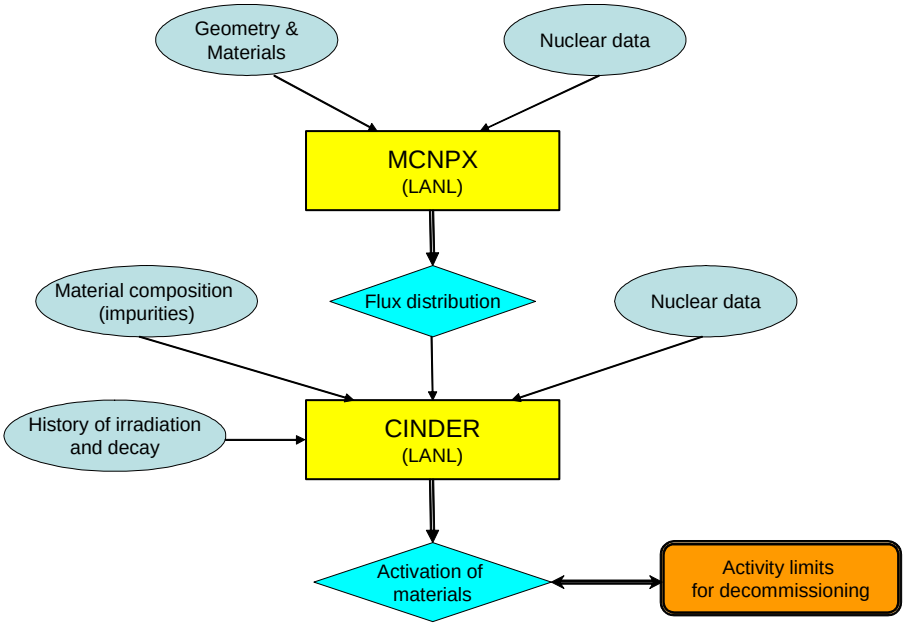

Figure 3: Neutron-flux distributions in the XY plane.

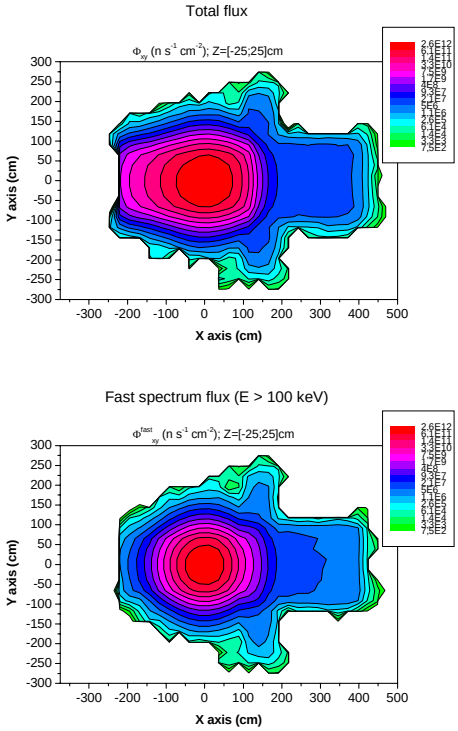

Figure 4: Activation of the first graphite column (Bq/g).

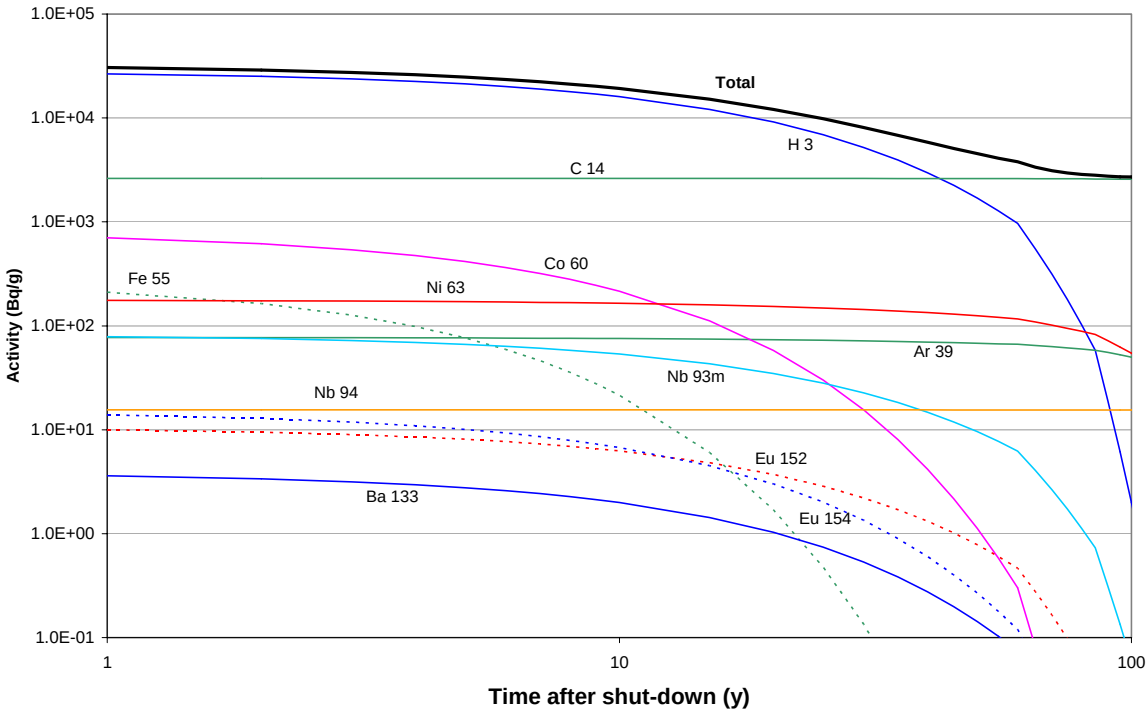

Table 1: Comparison of the neutron-flux (n/cm^2·sec).

	Experimental value		Calculation	Error
	Measure	Uncertainty		(C-E)/E
Central detector	1.27 E+12	± 5%	1.47 E+12	15.3 %
Outer detector	7.94 E+11	± 5%	9.38 E+11	18.2 %

Table 2: Comparison of the radioactivity of the graphite sample (Bq/g).

	Measured	Calculated
^{3}H *	54	82.69
^{14}C *	30	9.74
^{36}Cl		0.001
^{41}Ca		0.006
^{55}Fe	<15	0.47
^{60}Co	2.2	1.92
^{63}Ni	<10	0.65
^{133}Ba		0.012
^{134}Cs		0.017
^{152}Eu	<1.3	0.034
^{154}Eu	<0.9	0.039
Total		95.64

* Impurity levels of ^{6}Li and ^{14}N were assumed to be 0.15 and 200 ppm respectively.