Masterproef industriële ingenieurswetenschappen

Improving the Radionuclide Inventory Determination of the Irradiated Graphite from BR1 in Mol

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Introduction & Objective

In total, 492 tons of graphite acts as moderator and reflector for the BR1 (Belgian Reactor 1), located at the site of SCK·CEN (Studiecentrum voor Kernenergie) in Mol. Radionuclides in the graphite are produced due to nuclear reactions between neutrons and stable nuclies, mainly impurities in the graphite, since the start of the reactor in 1956. The objective of this work was to carry out a numerical simulation of the graphite activation in the BR1 to estimate the radionuclide inventory and specific activity at different locations in the graphite structure. The Monte Carlo burn-up code ALEPH was used for the complex calculation. The fuel configuration changes have been taken into account as well. Together with the current acceptance criteria for surface disposal, published by NIRAS/ONDRAF[1] there was discussed which volumes were suited for surface or geological disposal.

Figure 1: Spare blocks of BR1 graphite moderator

Methodology

The differential equation which describes the production and decay of ^{60}Co by neutron activation of ^{59}Co and the corresponding solution for the activity are illustrated below:

$$\frac{dN_{60_{Co}}(t)}{dt} = \sigma_{59_{Co}} . \varphi . N_{59_{Co}}(t) - \lambda_{60_{Co}} . N_{60_{Co}}(t)$$

$$A_{60_{Co}}(t) = \sigma_{59_{Co}} . \varphi . N_{1,0} . (1 - e^{-\lambda_{60_{Co}} t})$$

$N_{60_{Co}}$: Number of ^{60}Co atoms
$\sigma_{59_{Co}}$: Spectrum averaged microscopic cross section for (n, γ) reaction with 59Co
φ : Neutron flux (n/m².s)
$N_{59_{Co}}$: Number of 59Co atoms present at time t
$\lambda_{60_{Co}}$: Decay constant of ^{60}Co
$A_{60_{Co}}$: Activity (Bq) of ^{60}Co at time t
$A_{60_{Co}} = \lambda_{60_{Co}} . N_{60_{Co}}$
σ_{av} : Spectrum averaged cross section
$\sigma(E)$: Energy dependant reaction cross section
$\varphi(E)$: Energy dependant flux values, Calculated by MCNP
E_1, E_2 ; Energy group boundaries

The spectrum averaged cross section is calculated by ALEPH after the MCNP run. These energy independant reactions cross sections are used to calculate the reaction rates.

$$\sigma_{av} = \frac{\int_{E_1}^{E_2} \sigma(E)\varphi(E)d(E)}{\int_{E_1}^{E_2} \varphi(E)d(E)}$$

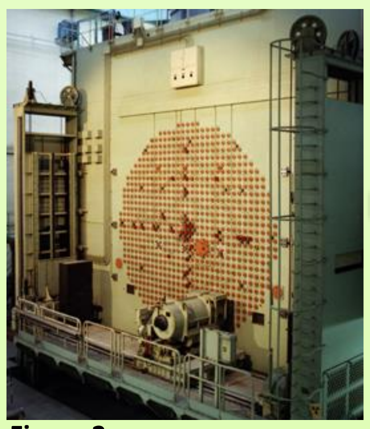
Reaction rate $R = \sigma_{av}{}^{1} . \varphi^{2} . N^{3}$ dependant on several parameters:

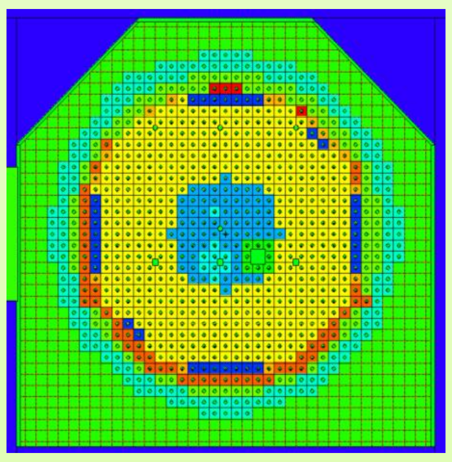
1: Position in the graphite pile

2: Position, $P_{thermal}$ of BR1 and fuel configuration

3: Initial element composition and concentration of the graphite. This parametesr has the major impact on the graphite activation. Values based on measurements and literature values. Obvious difference between graphite A & B quality.

For this problem, these sets of equations and corresponding solutions become very complicated (see Figure 3), therefore computercodes were used.





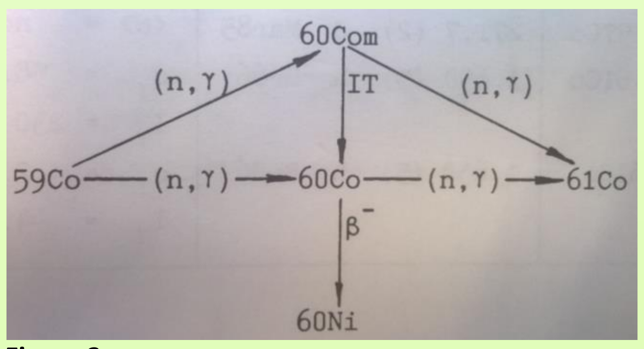
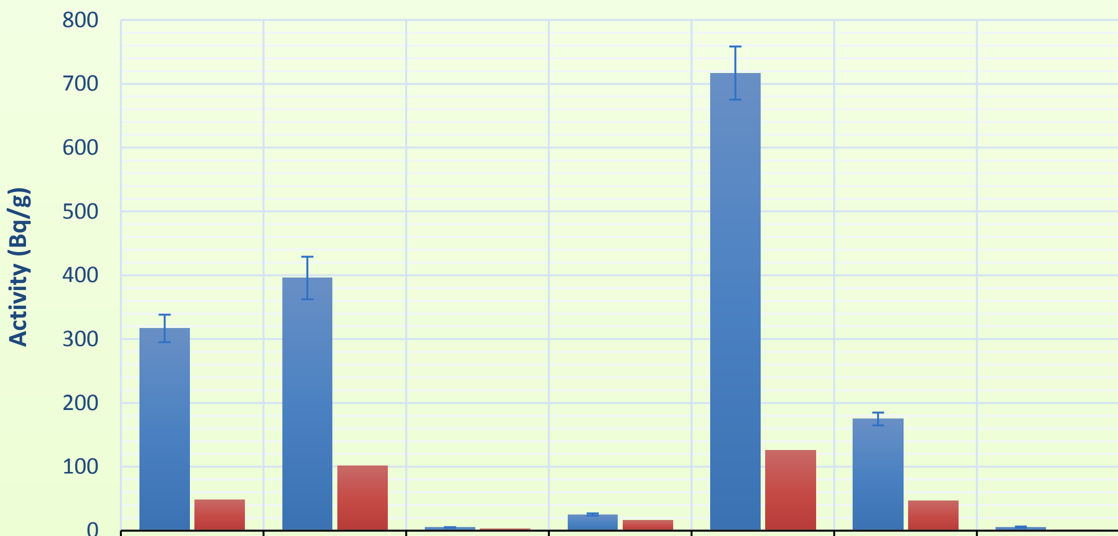


Figure 3: Production and decay scheme of ^{60}Co, starting from ^{59}Co with (n,γ) reaction

Figure 2: Left: Picture of BR1. Right: Front view of computer geometry of graphite matrix and fuel channels in BR1.

- The complete impurity list for graphite A & B was implemented in the model
- The loaded fuel channel configuration changes during the operation of the BR1 were implemented, starting at 501 channels loaded until the 552 current configuration.
- Many graphite samples were defined in the graphite stack at different locations, shown in Figure 6. This was done to see the impact of the place-dependant neutron fluxes and spectra as well as graphite A & B quality on the graphite activation.

Results



	60Co	65Zn	133Ba	134Cs	152Eu	154Eu	155Eu
Measurements	316,7	395,8	4,8	25,0	716,7	175,0	5,3
Simulation	48,21	101,72	2,77	16,02	125,87	46,92	1,31

Figure 4: Comparison of specific activity levels (Bq/g) between measurement and simulation for the measured radionuclides of sample E, located in graphite A of fuel channel C2.3.

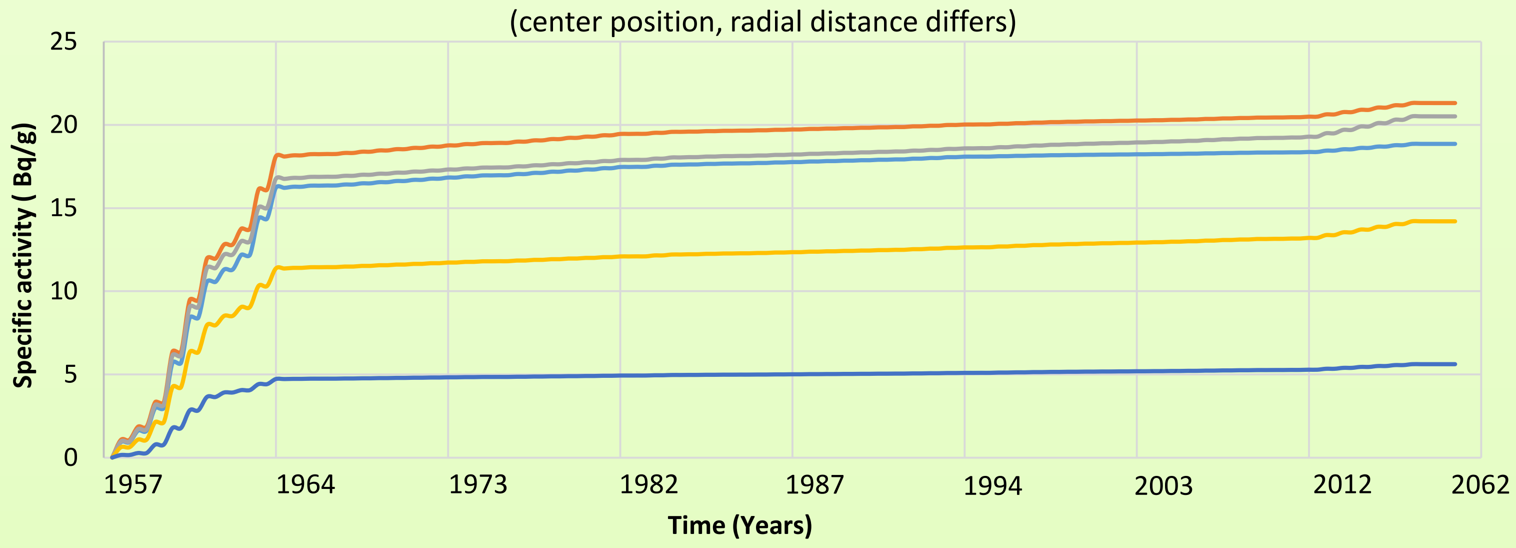


Figure 5: Evolution of ^{36}Cl production (Bq/g) in different graphite A locations from 1957 to 2012 and predicted up to 2062.

Channel	MATERIAL NUMBERS				DISPOSAL OPTION			
	0 cm	120 cm	240 cm	300 cm	0 cm	120 cm	240 cm	300 cm
A0.18	213	214	215	216	PSD	PSD	**SD**	**SD**
A0.16	209	210	211	212	PSD	PSD	PSD	PSD
A0.14	424	425	426	208	PSD	PSD	PSD	PSD
A0.11	421	422	423	207	PSD	PSD	PSD	PSD
A0.8	418	419	420	206	PSD	PSD	PSD	PSD
A0.6	415	416	417	205	PSD	PSD	PSD	PSD
A0.4	412	413	414	204	PSD	PSD	PSD	PSD
A0.2	409	410	411	203	PSD	PSD	PSD	PSD
A0.0	**406**	**407**	**408**	**202**	**PSD**	**PSD**	**PSD**	**PSD**
D1.3	403	404	405	201	PSD	PSD	PSD	PSD
C2.3	400	401	402	200	PSD	PSD	PSD	PSD

Figure 6: Channels and axial positions of the 44 graphite volumes where the activation calculation has been performed on at the end of 2012. The brown colour represents graphite A quality, while the red graphite B. Based on the X and Y criteria for radiological waste[1], the current simulation shows the graphite regions for surface disposal (SD) and possible surface disposal (PSD).

Discussion & Conclusion

The measurements show a big variation compared to the computer simulation. This result can be explained by the possible variance in impurity concentration between the individual graphite blocks of the same quality. It was impossible to prove this assumption because only one measurement was carried out on virgin graphite in the past. This show that the graphite composition, which was used in the computer model has the major impact on the graphite activation. Regarding the disposal option, all graphite volumes were destined for "possible surface disposal" except graphite material 215 and 216, which are located at the outside of the pile, in massive graphite B blocks. This result demonstrates that the 42 out of the 44 volumes use more radiological capacity than an average waste package (Y criterion value > 1). The dominant radionuclides for the Y criterion value were ^{36}Cl and ^{94}Nb.

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References: [1] Surface waste disposal acceptance criteria, NIROND-TR 2007-05,2008

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