CARBOWASTE

Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste

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Technical Report T-3.3.3-b

Characterization of Irradiated BR1 Graphite

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Document title
Characterization of Irradiated BR1 Graphite **Technical Report**

Executive summary

An investigation has been performed of the radionuclide inventory generated due to activation of the graphite reflector and moderator of the BR1 research reactor of SCK•CEN.
The radionuclides studied were in particular C-14, Cl-36 and Co-60, all of them being β-emitters. The approach followed was through theoretical calculations using the Origen 2.2 code. The activity of radioisotopes was calculated for several periods of time taking into account the irradiation history of BR1 graphite.

We present the results of the calculations for different scenario's for the concentration of impurities in the virgin graphite.

This is the contribution of SCK•CEN to Task 3.3: Localisation of Impurities and Isotopes before and after Treatment

Revisions

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00	dd/mm/yyyy	Issue	Name, Organisation	Name, Organisation *Signature*	Name, Organisation *Signature*	Name, Organisation *Signature*
01	27/03/2013	First issue	Van Iseghem SCK•CEN	Van Hecke SCK•CEN	Pina CIEMAT	von Lensa FZJ

1. Objective of the study

An investigation has been performed of the radionuclide inventory generated due to activation of the graphite reflector and moderator of the BR1 research reactor of SCK•CEN. The objective was to obtain data on the radionuclide inventory, in view of a future evacuation and disposal of the graphite radioactive waste.

The radionuclides were generated due to activation of the graphite material, and the impurities included in the graphite. The radionuclides studied were in particular C-14, Cl-36 and Co-60, all of them being β-emitters. The main approach followed was through theoretical calculations using the Origen 2.2 code. The activity of radioisotopes was calculated for several periods of time taking into account the irradiation history of BR1 graphite.

We briefly list the reactions that can generate the radionuclides of concern:

- Cl-36 (3.01 x 10^5 years half life):
    - $35Cl(n,\gamma)Cl36$
    - $39K(n,\alpha)Cl36$
    - $34S(n,\gamma)S35$ and then $35S(n,\beta^-)Cl35$

    The dominant route usually is by activation of Cl-35.

- C-14 (5,730 years half life):
    - $14N(n,p)C14$
    - $13C(n,\gamma)C14$
    - $17O(n,\alpha)C14$

The predominance of any of these reactions will depend on the concentration of the stable N-14, C-13 and O-17 isotopes in the virgin graphite.

- Co-60 (5.27 years half-life):
    - $58Fe(n,\gamma)Fe59$, $59Fe(-\beta)Co59$ and $59Co(n,\gamma)Co60$

A detailed description of the results can be found in [1].

2. The BR1 reactor

The BR1 reactor is the first research reactor built in Belgium. It reached its first criticality in May 1956 and it has been working till these days. BR1 is placed at the facilities of SCK.CEN located in Mol (Belgium).

BR1 is a natural uranium reactor moderated by graphite and cooled by means of air which flows inside the fuel channels. The moderator is graphite class A, its density is 1,72 g/cm^3 and includes 0,4 ppm of boron traces. Beyond a radius of 2,6 m graphite of class B is placed as a reflector. Its density is 1,62 g/cm^3 and contains 1,6 ppm boron traces and other impurities such as Fe, Al, K, Ca, Ni and V. The total mass of graphite in the BR1 reactor is about 500 tons.

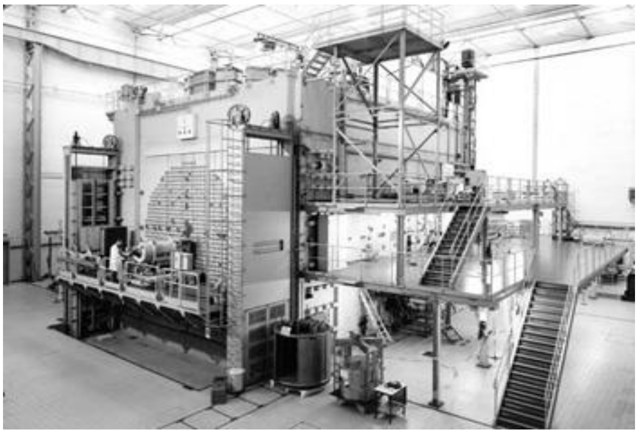

Figure 1: view of the BR1 reactor (ref 49)

Figure 2 shows a cross section of the reactor, showing the different parts of the reactor.

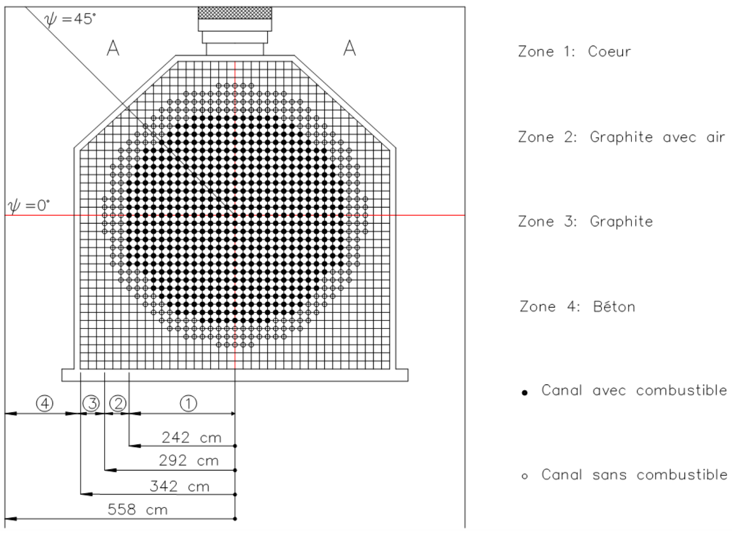

Figure 2: cross section of the BR1, showing the different main components.

3. Methodology

The 1D - Origen 2.2 neutron activation code was used to calculate the quantity of activation products generated during the irradiation in the BR1 reactor. Input data for the calculations were the cooling time (after irradiation), the concentration of the impurities in the graphite, the neutron flux (which is a function of time and position), and the type of graphite.

The concentration of the impurities in both (unirradiated) graphite type A and B was determined with ko- NAA (neutron activation analysis). Table 3 lists the values for the impurities that are precursors for Cl-14, Cl-36 and Co-60:

Table I: Concentration of main impurities in graphite, as measured by k0-NAA.

Element	Graphite A	Graphite B
S mg/Kg	< 20	< 300
Cl mg/Kg	1,82 ± 0,1	4,8 ± 0,2
K mg/Kg	1,53± 0,04	5,6 ± 0,2
Co ng/g	10 ± 2	99 ± 4

For some other consituents such as N and Li, as no k0-NAA measurements were available, we made estimations based on literature data.

Other parameters considered in this study were:

- The variation of the neutron flux with the radial distance was accounted for using the figures 3 (graphite type A) and 4 (graphite type B). These plots were calculated taking into account the evolution of the spatial neutron flux over the years of operation of the reactor. Details can be found in [2].

- The various scenarios considered in order to determine the effect of impurities in the unirradiated graphite are reported in the next section, together with the results of the calculations of the activation products.

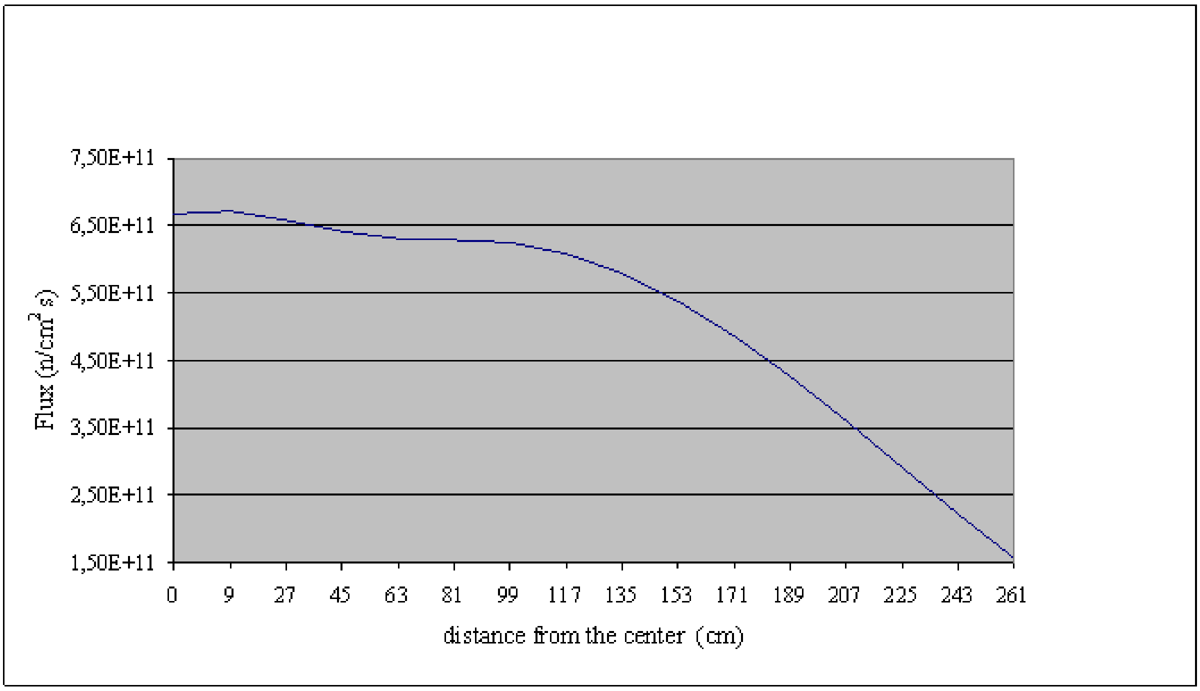

Figure 3: Flux variation in graphite type A [2].

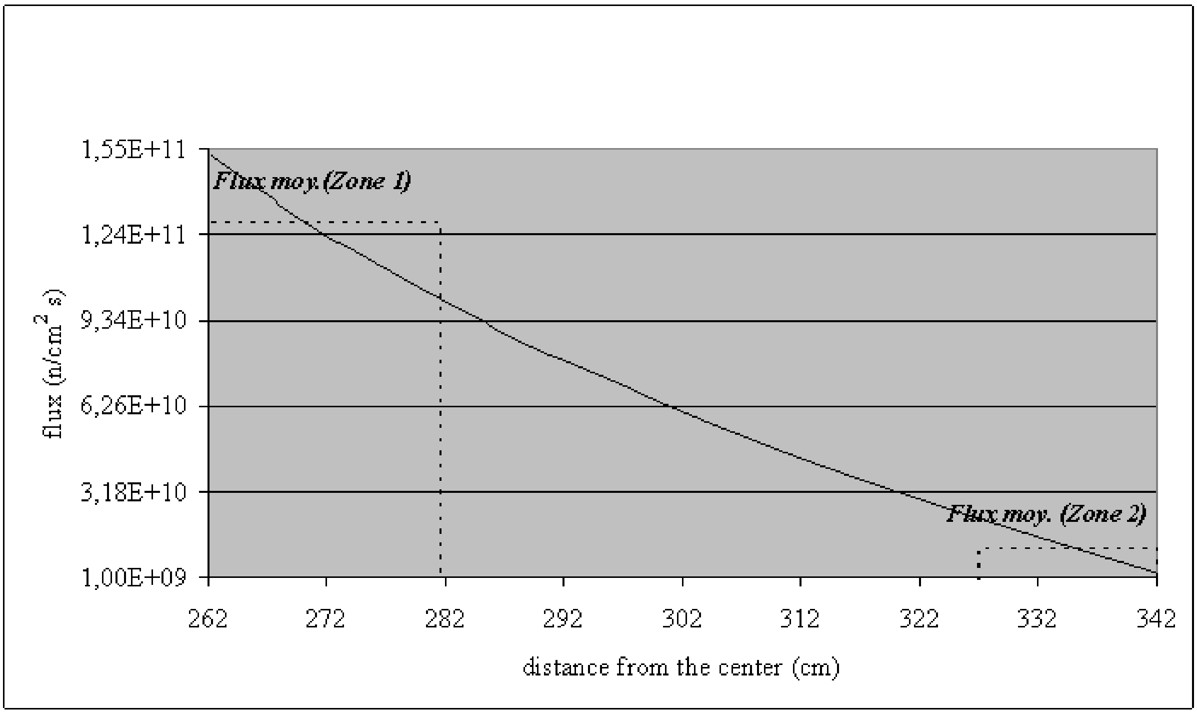

Figure 4: Flux variation in graphite type B [2].

4. Scenario's considered, and results of the calculations

Graphite type A

Scenario 1: Base concentration without nitrogen and lithium, other constituents based on k0-NAA. The objective is to determine the base activity for graphite type A.

Scenario 2: Base concetration with 10 ppm of nitrogen and 0,05 ppm of lithium. The objective it to determine the variation of C-14 and H-3 activity.

Scenario 3: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium and 0,1 ppm of Co. The objective is to determine the variation of Co-60 activity.

Scenario 4: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium and 0,5 ppm of Co. The objective is to determine the variation of Co-60 activity as a function of its concentration.

Scenario 5: Base concentration is doubled for sulfur, potassium, calcium, iron, cesium and europium. Also, graphite includes 10 ppm of nitrogen and 0,05 ppm. The objective is to determine the variation of Cl-36, Ca-14, Fe-55, Ni-63, Cs-134, Eu-152 and Eu-154 activities.

Scenario 6: Base concentration is quadrupled for sulfur, potassium, calcium, iron, cesium and europium. Also, graphite includes 10 ppm of nitrogen and 0,05 ppm. The objective is to determine the variation of Cl-36, Ca-14, Fe-55, Ni-63, Cs-134, Eu-152 and Eu-154 activities and their sensitivity with regard their concentration.

Scenario 7: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium and the concentration of chlorine is doubled. The objective is to see the effect of the increment of chlorine in the Cl-36 activity.

Below we show some plots for different scenario's.

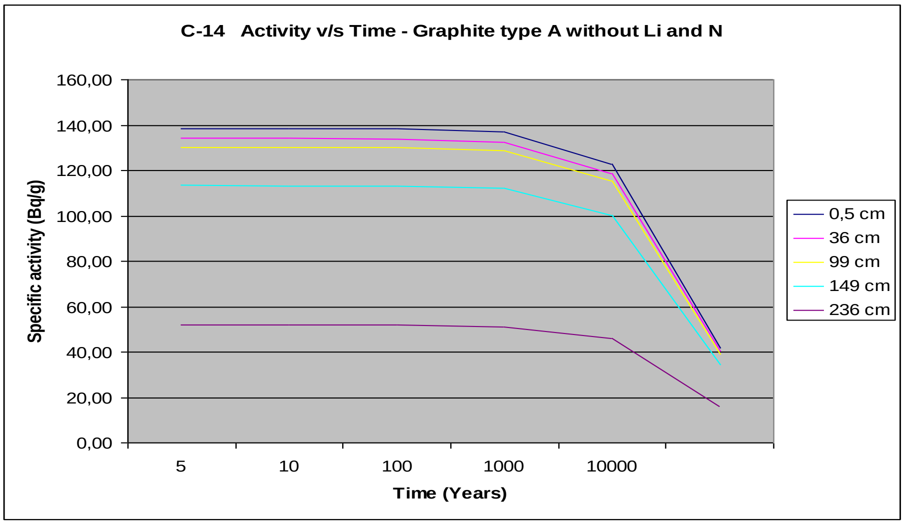

Figure 5: Graphite type A, C-14 activity evolution for different distances from reactor center, without lithium and nitrogen.

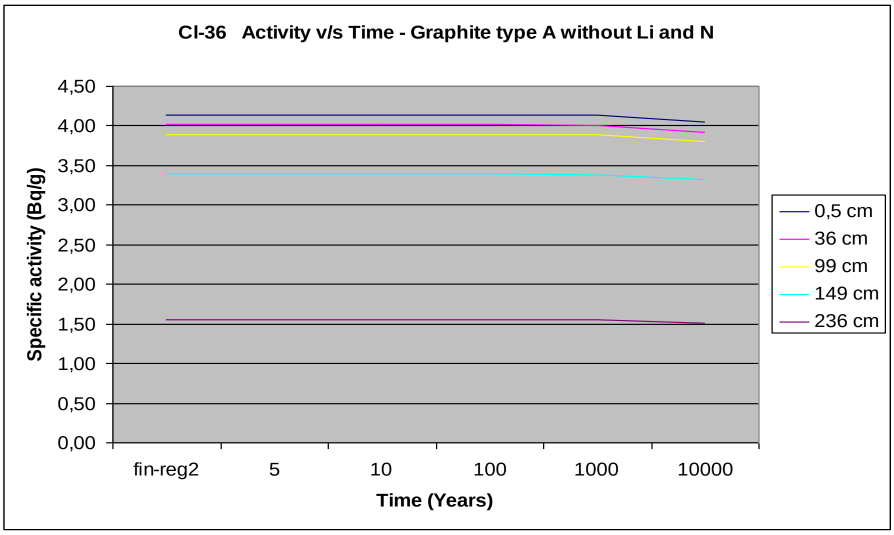

Figure 6: Graphite type A, Cl-36 activity evolution for different distances from reactor center, without lithium and nitrogen.

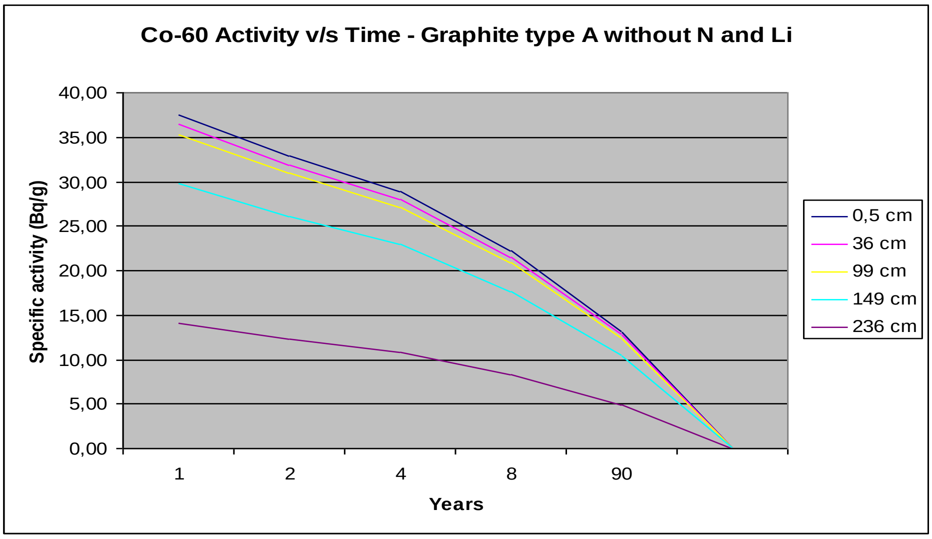

Figure 7: Graphite type A, Co-60 activity evolution for different distances from reactor center, without lithium and nitrogen.

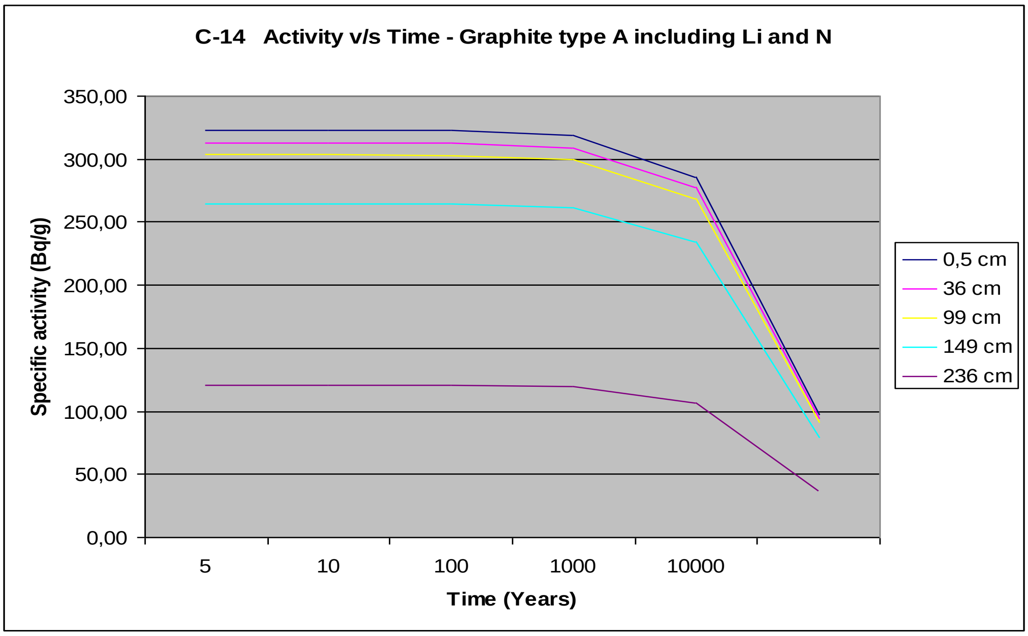

Figure 8: Graphite type A, C-14 activity evolution for different distances from reactor center, including lithium and nitrogen.

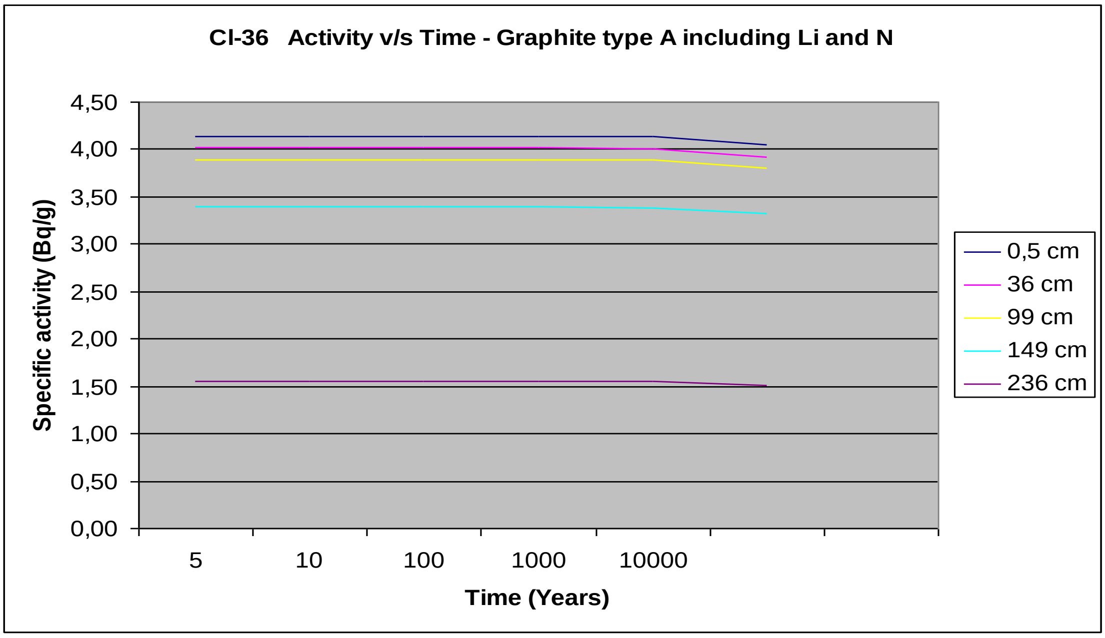

Figure 9: Graphite type A, Cl-36 activity evolution for different distances from reactor center, including lithium and nitrogen.

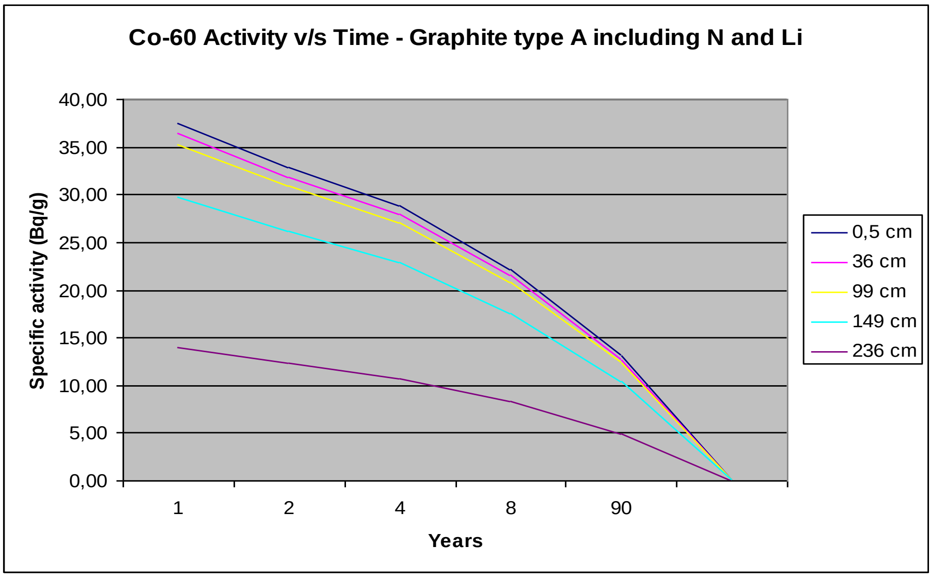

Figure 10: Graphite type A, Co-60 activity evolution for different distances from reactor center, including lithium and nitrogen.

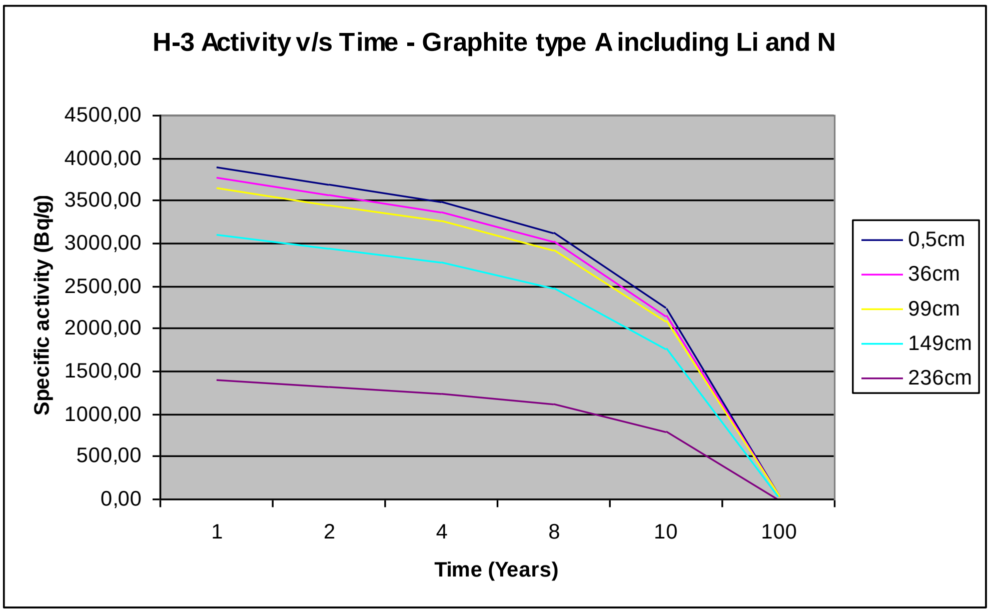

Figure 11: Graphite type A, H-3 activity evolution for different distances from reactor center, including lithium and nitrogen.

Graphite type B

Scenario 8: Base concentration without nitrogen and lithium, other constituents based on k0-NAA. The objective is to determine the base activity for graphite type B.

Scenario 9: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium. The objective it to determine the variation of C-14 and H-3 activity.

Scenario 10: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium and 0,1 ppm of Co. The objective is to determine the variation of Co-60 activity.

Scenario 11: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium and 0,5 ppm of Co. The objective is to determine the variation of Co-60 activity.

Scenario 12: Base concentration is doubled for sulfur, potassium, calcium, iron, cesium and europium. Also, graphite includes 10 ppm of nitrogen and 0,05 ppm. The objective is to determine the variation of Cl-36, Ca-14, Fe-55, Ni-63, Cs-134, Eu-152 and Eu-154 activities.

Scenario 13: Base concentration is quadrupled for sulfur, potassium, calcium, iron, cesium and europium. Also, graphite includes 10 ppm of nitrogen and 0,05 ppm. The objective is to determine the variation of Cl-36, Ca-14, Fe-55, Ni-63, Cs-134, Eu-152 and Eu-154 activities and their sensitivity with regard their concentration.

Scenario 14: Base concentration with 10 ppm of nitrogen and 0,05 ppm of lithium and the concentration of chlorine is doubled. The objective is to see the effect of the increment of chlorine in the Cl-36 activity.

Below we show some plots for different scenario's.

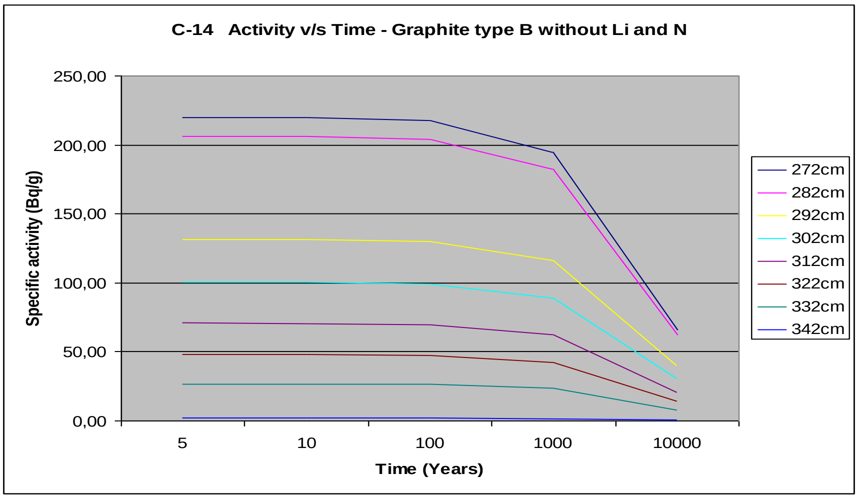

Figure 12: Graphite type B, C-14 activity evolution for different distances from reactor center, without lithium and nitrogen.

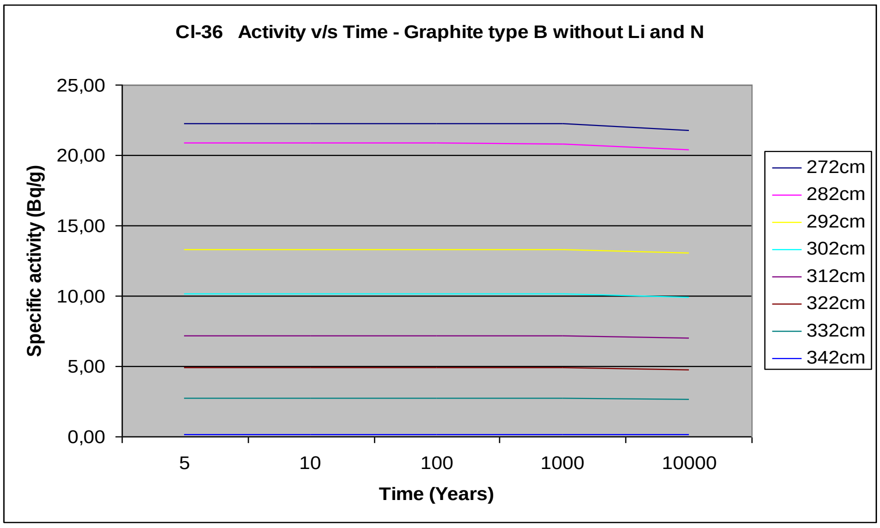

Figure 13: Graphite type B, Cl-36 activity evolution for different distances from reactor center, without lithium and nitrogen.

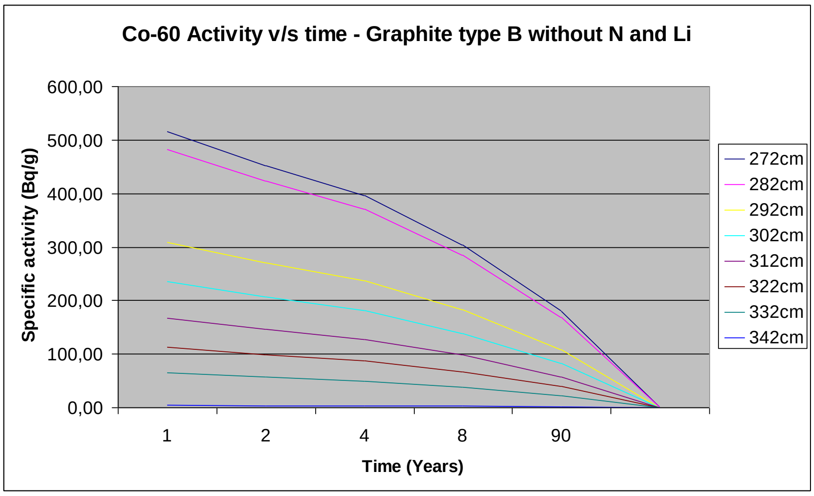

Figure 14: Graphite type B, Co-60 activity evolution for different distances from reactor center, without lithium and nitrogen.

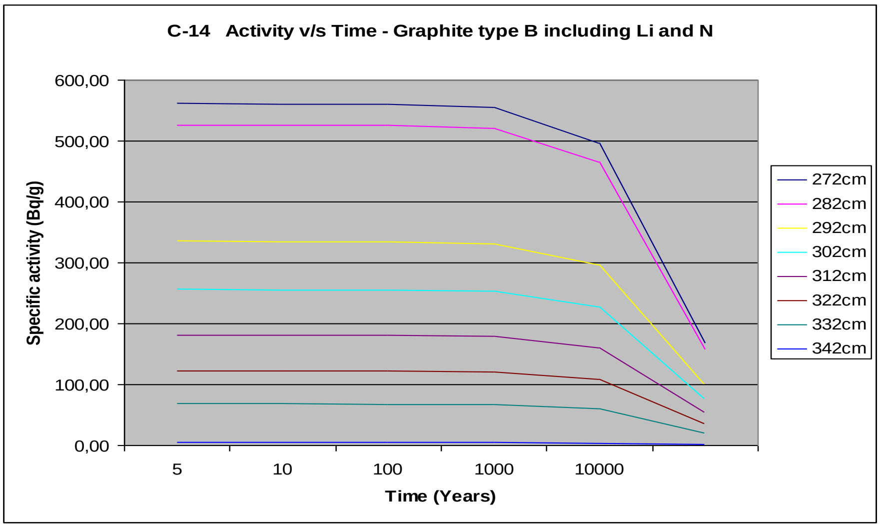

Figure 15: Graphite type B, C-14 activity evolution for different distances from reactor center, including lithium and nitrogen.

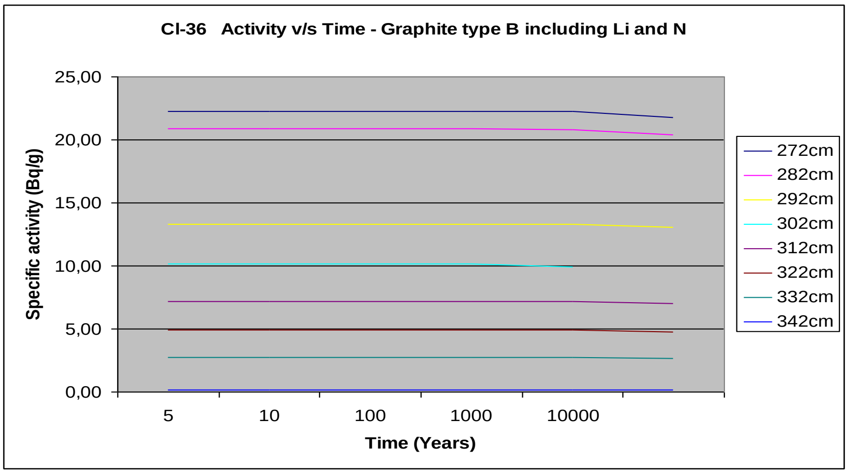

Figure 16: Graphite type B, Cl-36 activity evolution for different distances from reactor center, including lithium and nitrogen

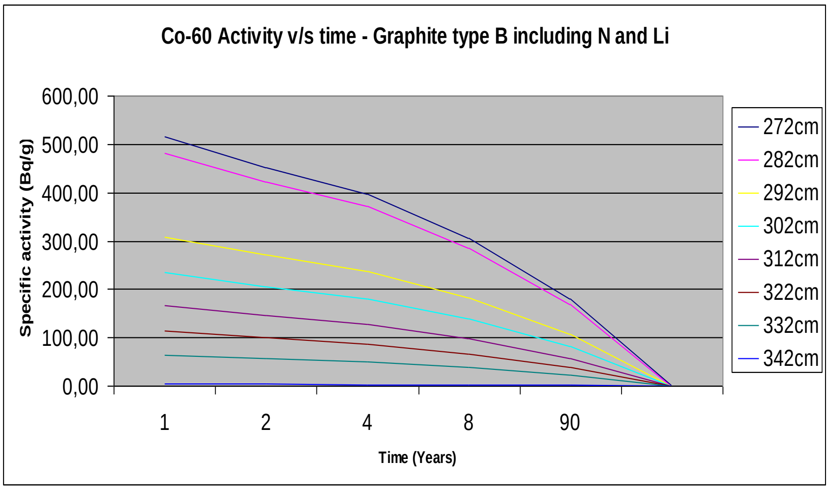

Figure 17: Graphite type B, Co-60 activity evolution for different distances from reactor center, including lithium and nitrogen.

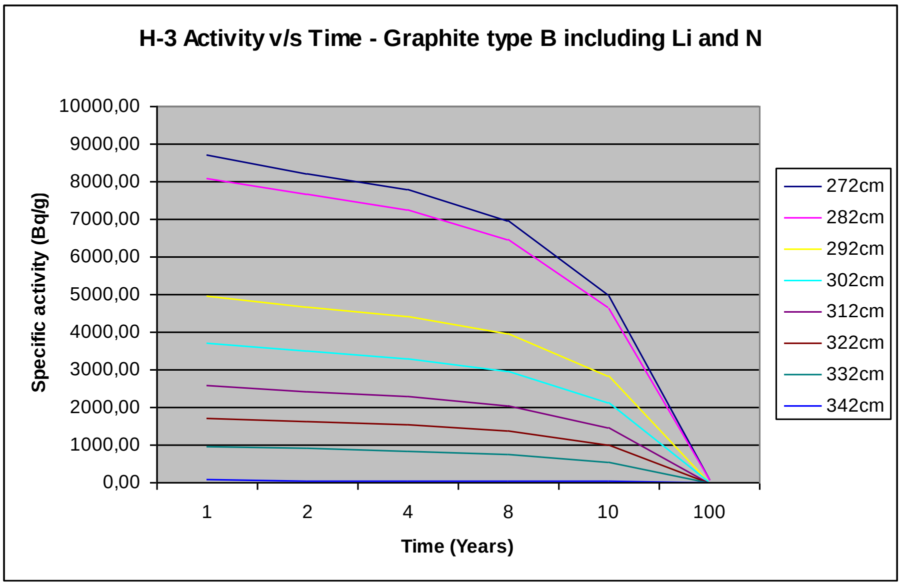

Figure 18: Graphite type B, H-3 activity evolution for different distances from reactor center, including lithium and nitrogen.

5. Discussion, conclusions

The results of the calculations were used to evaluate the effect of the impurities present in the virgin graphite, see Tables II for graphite type A and Tables III for graphite type B. Note, these calculations are performed at the positions in the graphite closest to the core of the reactor: 0.5 cm in case of graphite A, 272 cm in case of graphite B.

Graphite type A:

Table II-a: Nitrogen and Lithium influence on C-14 and H-3 activities in graphite A.

	Graphite type A	
	Scenario 1 (0 ppm N / 0 ppm Li)	Scenario 2 (10 ppm N / 0,05 ppm Li)
Scenario total activity (Bq/g)	1.082	5.191
C-14 (Bq/g)	138	322,6
H-3 (Bq/g)	1,85	3899

Table II-b: Cobalt influence on Co-60 activity in graphite A.

	Graphite type A	
	Scenario 3 (0,1 ppm of Co)	Scenario 4 (0,5 ppm of Co)
Scenario total activity (Bq/g)	5.524	7.000
Co-60 (Bq/g)	369,8	1.847

Table II-c: Impact of scenarios 5 and 6 in graphite A.

	Graphite type A		
	Scenario 1	Scenario 5	Scenario 6
Scenario total activity (Bq/g)	5.191	5.901	7.326
Cl-36 (Bq/g)	4,14	4,15	4,16
Ca-41 (Bq/g)	3,44	6,87	13,74
Fe-55 (Bq/g)	38,9	77,96	155,92
Ni-63 (Bq/g)	235,58	471	942
Cs-134 (Bq/g)	30	61,2	123
Eu-152 (Bq/g)	279,94	559,8	1.119,6
Eu-154 (Bq/g)	31,96	63,8	127,5

Table II-d: Chlorine influence on Cl-36 and S-35 activities in graphite A.

	Graphite type A	
	Scenario 1 (1,8 ppm of Cl)	Scenario 7 (3,6 ppm of Cl)
Total activity (Bq/g)	5.191	5.218
Cl-36 (Bq/g)	4,14	8,28
S-35 (Bq/g	28,77	51,28

Graphite type B:

Table III-a: Nitrogen and Lithium influence on C-14 and H-3 activities in graphite B.

	Graphite type B	
	Scenario 8 (0 ppm N / 0 ppm Li)	Scenario 9 (10 ppm N / 0,05 ppm Li)
Total activity (Bq/g)	8.558	17.630
C-14 (Bq/g)	220	561
H-3 (Bq/g)	0	8.702

Table III-b: Cobalt influence on Co-60 activity in graphite B.

	Graphite type B	
	Scenario 10 (0,1 ppm of Co)	Scenario 11 (0,5 ppm of Co)
Total activity (Bq/g)	17.630	19.717
Co-60 (Bq/g)	520	2.603

Table III-c: Impact of scenarios 5 and 6 in graphite B.

	Graphite type B		
	Scenario 9	Scenario 12	Scenario 13
Total activity (Bq/g)	17.630	27.054	41.773
Cl-36 (Bq/g)	22,9	22,9	22,9
Ca-41 (Bq/g)	40,7	80,14	160
Fe-55 (Bq/g)	400	800	1600
Ni-63 (Bq/g)	2.475	4.950	9.901
Cs-134 (Bq/g)	28	56,35	112
Eu-152 (Bq/g)	3.265	6.530	13.061
Eu-154 (Bq/g)	348	695,9	1.391

Table III-d: Chlorine influence on Cl-36 and S-35 activities in graphite B.

	Graphite type B	
	Scenario 9 (4,8 ppm of Cl)	Scenario 14 (9,6 ppm of Cl)
Total activity (Bq/g)	17.630	17.753
Cl-36 (Bq/g)	22,2	44,5
S-35 (Bq/g)	289,3	389,6

We can draw a few conclusions from the Tables II and III:

- Tables II-a and III-a show that the formation of C-14 and H-3 is mainly the result of the activation of N-14 (reaction 14N(n,p)C14)) and Li-6 (reaction 6Li(n,α)H3)), respectively.
- Tables II (c,d) and III (c,d) show that Cl-36 is almost solely formed by activation of Cl-35 (35Cl(n,γ)Cl36), not by activation of S.
- The impact of the higher concentration of impurities in graphite B compared to graphite A on the generation of activation products is stronger than the impact of the higher neutron flux for graphite A.

6. Conclusions, future work

The work presented had offerred a first attempt to calculate the inventory of activation products in the BR1 reactor. It will enable already to have a first judgement on the inventory of long-living radionuclides (especially Cl-36 and C-14) generated.
It is intended to continue this work as follows:

- Samples of the irradiated graphite will be taken, and analyzed radiochemically. The objective is to compare with the calculated radionuclide concentrations, and therefore to validate the predicted inventories.
- 2 or 3D calculations are planned using a coupling between the MCNP (Monte Carlo) code and the ORIGEN 2.2 activation code, as an improvement of the modeling predictions.
- The results of the radionuclide inventory calculations will be compared with the acceptance criteria for surface disposal in Belgium.

7. Bibliography

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[2] N.Messaoudi. “Caractérisation radiologique des matériaux de structure du BR1- Evaluation de l’activité des matériaux de estructure de BR1”.Passif technique. SCKCEN, 2000.