EUROPEAN COMMISSION

Community Research

SEVENTH FRAMEWORK PROGRAMME

CARBOWASTE

Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste

Grant Agreement Number: FP7-211333

Technical Report T-3.4.1
Assessment of isotope-accumulation data from MTR

Authors: Daniela Gugiu, Csaba Roth

Reporting period: 01/04/2008– 31/03/2011

Date of issue of this report : 20/05/2011

Start date of project : **01/04/2008** Duration : **48 Months**

Project co-funded by the European Commission under the Seventh Framework Programme (2007 to 2011) of the European Atomic Energy Community (EURATOM) for nuclear research and training activities		
Dissemination Level		
PU	Public	
RE	Restricted to the partners of the CARBOWASTE project	**X**
CO	Confidential, only for specific distribution list defined on this document	

Distribution list

Person and organisation name and/or group	Comments
Werner von Lensa, FZJ	
Gabriel Pina, CIEMAT	
Abbie Jones, UoM	

CARBOWASTE		
Work package: 3 **Task: 3.4**	**CARBOWASTE document no:** CARBOWASTE-1104-T-3.4.1	**Document type:** T=Technical Report
Issued by: INR (RO) **Internal no.:** 9378/2011		**Document status:** Final

Document title

Assessment of isotope-accumulation data from MTR

Executive summary

The changes observed in the graphite structure and in the content of impurities after irradiation depend strongly on the irradiation history of the material. For this reason it is necessary to have accurate information about the neutron flux-spectrum and the initial content of the impurities. The MCNP5 code is used in the present study in order to evaluate the neutron flux-spectrum in the graphite through a detailed simulation of the actual geometry and composition of the thermal column and TRIGA reactor. The neutron flux-spectrum is subsequently used in the FISPACT code which provided the graphite isotopes inventory at various irradiation and cooling time steps.

Revisions

Rev.	Date	Short description	Authors	Internal Review	Task Leader	WP Leader
01	20/04/2011	Issue	Daniela Gugiu INR	Csaba Roth INR	Crina Bucur INR *Signature*	Daniela Diaconu INR *Signature*

Table of Content

1 Generals

The thermal column of TRIGA reactor consists in a block of graphite formed by 96 rectangular bricks of graphite encased in Aluminum. The thermal column is placed on a stainless steel frame fixed in the reactor pool, in the northern part of the steady state zone [1].

TRIGA 14 MW is a double core material testing reactor having a Steady State Reactor - TRIGA SSR (14MW), and an Annular Core Pulsing Reactor -TRIGA ACPR. In the active zone of SSR, the neutron fluxes range from 2.7 10^{13} to 3.2 10^{14} neutrons/cm^{2}s, while in the thermal column, the density of the neutron flux can be 2.0 10^{6} - 5.0 10^{10} neutrons/cm^{2}s.

2 Design of the thermal column

The thermal column having 12 rows of 8 bricks is 172x114x177cm has the geometry shown in Figure 1. Thermal column of the TRIGA reactor

.

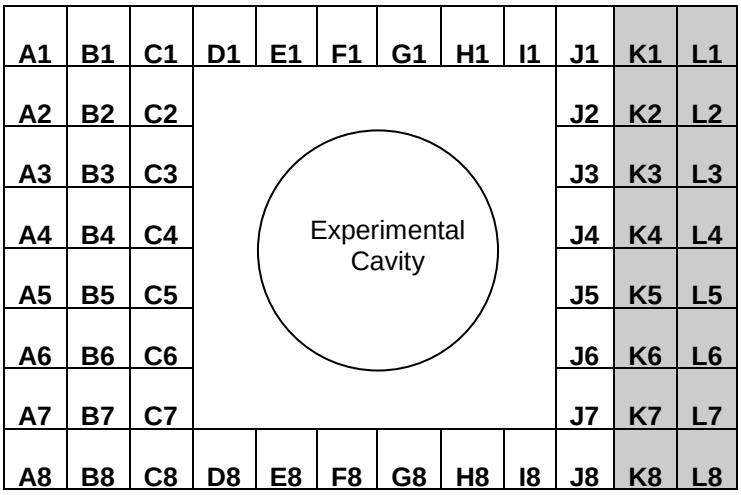

Figure 1. Thermal column of the TRIGA reactor

The graphite blocks have a square section of 14.3 cm length. The graphite used in TRIGA thermal column is sintered graphite produced in UK in the ’50 with a density of 1.63g/cm^3. The total graphite in a brick is 23.64 Kg and the whole column weights 2.3t.

2.1 Experimental data

Impurities analysis experimentally determined under WP 3.2 by complementary techniques (EDS, XRF) led to the following content (Table 1):

Table 1 Impurities content in Virgin graphite measured by XRF

Element	Percent wt (%)	Standard error
C	99.87	
Na	0.0628	0.0033
Ba	0.0229	0.0085
Si	0.0173	0.001
Mg	0.0070	0.0009
Cl	0.0063	0.0007
Ca	0.0038	0.0005
S	0.0031	0.0003
Th	0.0023	0.0009
Dy	0.0021	0.0008
K	0.0018	0.0005
Al	0.0011	0.0003
P	0.0011	0.0003

The sensitivity limit of XRF did not allow the identification of Co and Eu. Some previous activity measurements by gamma spectroscopy indicated that the concentration for both elements could be around 1 ppb.

3 Model of C-14 accumulation in MTR

3.1 Previous C-14 production rate estimation

In some earlier computations the C-14 production (yield) rate was estimated using MCNP-4C [2], based on the model of the thermal column illustrated in Figure 2.

At that time the results were reported to the reactor power based on the flux measurements performed in the central cavity of the thermal column which was created by replacing 36 central bricks with a graphite block having a spherical central cavity. A dry channel (8 m high) was developed on the top of the thermal flux cavity for access and instrumentation purposes.

The device is designed so that it allows the placement of the ΣΣ standard system and of the fission chambers afferent instrumentation into the cavity [3]. In that stage, the model assumed that C-14 arises only from neutron capture in C-13. Due to the long lifetime, all C-14 yielded during reactor operation accumulates.

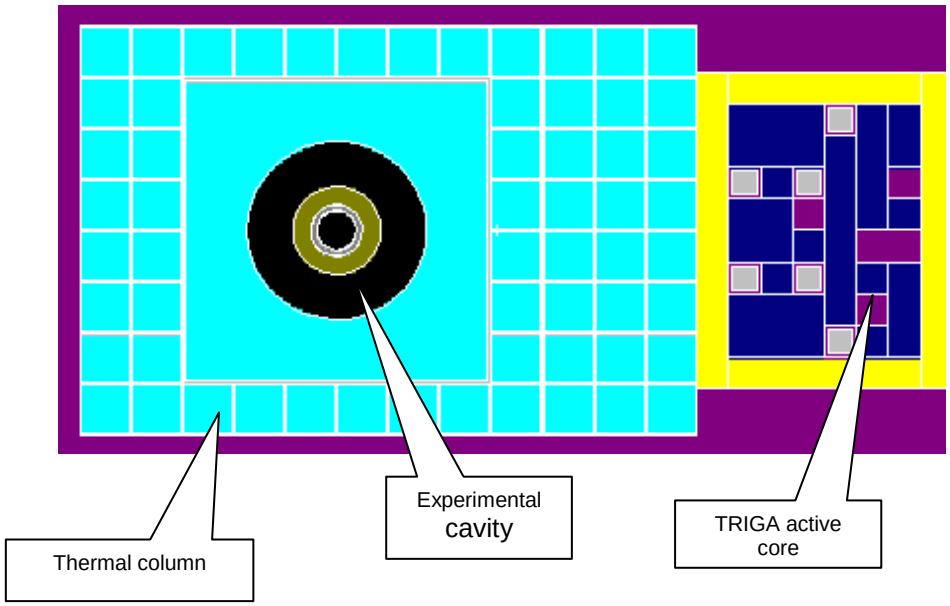

Figure 2 MCNP model of the facilities

The main outputs of the MCNP-4C simulation have been:

- neutron flux density in the center of the experimental cavity – Φ - needed to scale the results to the reactor power
- C-14 yielding rate – Ψ - averaged on the whole graphite volume of the thermal column

Based on the results carried out by the Monte Carlo simulation and taking into consideration a reference value of neutron flux density in the cavity center of 5 10^9 n/cm^{2}s at 1MW$_{th}$ [4] the corresponding reaction rate was:

Ψ=2.11 10^7ips/kg graphite

which means 1.82 10^{12} C-14 nuclei/MWday/kg graphite or 7 Bq/ MWday/kg graphite.

As a result, the estimated C-14 activity in the whole volume of the thermal column consisting in 98 bricks was:

Λ=16.2 KBq/MW day,

and the average value of the neutron flux density in the whole thermal column corresponding to 1MW$_{th}$ (thermal power) was of 4.7 10^{10} n/cm^{2}s.

3.2 Isotopes accumulation in MTR graphite

Some new evaluations of the isotopes accumulation in the thermal column graphite, including the C-14, have been performed using the MCNP5 and the FISPACT [5] codes.

As first step, criticality computations using MCNP5 code have been performed taking into account the detailed geometry and composition of the thermal column and of the TRIGA reactor active core (as previously) in order to estimate the average neutron flux in the thermal column; this time, the thermal $S(\alpha,\beta)$ neutron cross-section library (for graphite) has been used leading to a better estimation of the neutron flux-spectrum in the thermal column.

The power normalization factor (FP) of the MCNP5 evaluated neutron flux is computed using the following formula:

$$FP = \frac{\nu P}{k_{\text{eff}}\, 1.602 \cdot 10^{-13} Q_{av}},$$

where ν is the average number of neutrons emitted per fission which is computed and provided in the output of MCNP5 code, P is the total reactor power (10 MWth), and Q_{av} represents the average value of the recoverable energy per fission. As a result, the absolute value of the average neutron flux in the whole thermal column is of $7.4 * E^{11}$ n/cm^2/s.

The second step consists in the evaluation of the graphite isotope inventory at various irradiation and cooling steps using the FISPACT code.

Taking into account that a detailed irradiation history is not available yet, it has been assumed that an irradiation time interval of 10 years at a reactor power of 10 MW$_{\text{th}}$ could be conservative. It should be mentioned that the total irradiation time (10 years) has been divided in smaller time steps in order to obtain more accurate results taking into account the presence of many short-lived nuclides; additionally, the values of some parameters required in the FISPACT input (as *LEVEL C N*) have been set appropriately. This option allows the input of the two parameters which determine the nuclides that are in equilibrium (*C*) and the number of time subintervals (*N*) into which the irradiation time is divided. It means that the nuclides with decay constant λ (s^{-1}) will be in equilibrium if $\lambda T > C$; also, the time subintervals (*N*) has been set to *N=10* since the rate of fission of actinides is only updated at the end of each subinterval and the Th is present as impurity in graphite.

Three types of FISPACT runs have been performed:

a) for pure graphite

b) for graphite with impurities in the amounts presented in Table 1 and obtained by XRF measurements; additionally, 1ppm of Co has been considered.

c) case b) plus N (1 ppm) and Eu (2 ppb) as impurities.

The activities (expressed in Bq/kg) of some isotopes of interest after 10 years of irradiation at constant power of 10 MW_{th} are presented in Table 2.

Table 4 presents the activities of the isotopes after 10 years of cooling.

Table 2. Isotope accumulations after 10 years of irradiation

Element	Case a) Activity (Bq/kg)	Case b) Activity (Bq/kg)	Case c) Activity (Bq/kg)
C-14	4.800E+03	4.794E+04	4.879E+04
Co-60	-	9.095E+04	9.095E+04
Eu-152	-	4.145E-02	1.304E+04
Eu-154	-	7.780E-01	1.841E+04
Cs-137	-	8.746E+04	8.746E+04
Cl-36	-	5.070E+02	5.070E+02
H-3	6.083E-02	3.068E+02	1.614E+05

C-14 activity

One could observes from Table 2 (case b) and c)) that the presence of various impurities in the graphite leads to an increase of the **C-14** activity relative to the case a) (pure graphite). As previously mentioned, a small amount (1 ppm) of N has been considered as impurity in the case c).

Co-59 activity

The amount of **Co-59** impurity considered in the simulations is of 1 ppm, leading to an activity of *2.442E+04 Bq/kg* or *24.42 Bq/g* after 10 years of cooling (Table 3).

Table 3. Isotope activities after 10 years of cooling

Element	Case a) Activity (Bq/kg)	Case b) Activity (Bq/kg)	Case c) Activity (Bq/kg)
C-14	4.794E+03	4.788E+04	4.873E+04
Co-60	-	2.442E+04	2.442E+04
Eu-152	-	2.496E-02	7.812E+03
Eu-154	-	3.608E-01	8.216E+03
Cs-137	-	1.551E+05	1.551E+05
Cl-36	-	5.070E+02	5.070E+02
H-3	3.467E-02	4.202E+02	9.223E+04

As could be seen in Figure 3 the measured activities of the Co-60 in various samples of the same graphite block vary from tens of Bq/g up to 100 Bq/g. This spread of the values leads to the conclusion that the Co impurities distribution in graphite is strongly non-uniform and it is difficult to perform an accurate comparison with the simulation results.

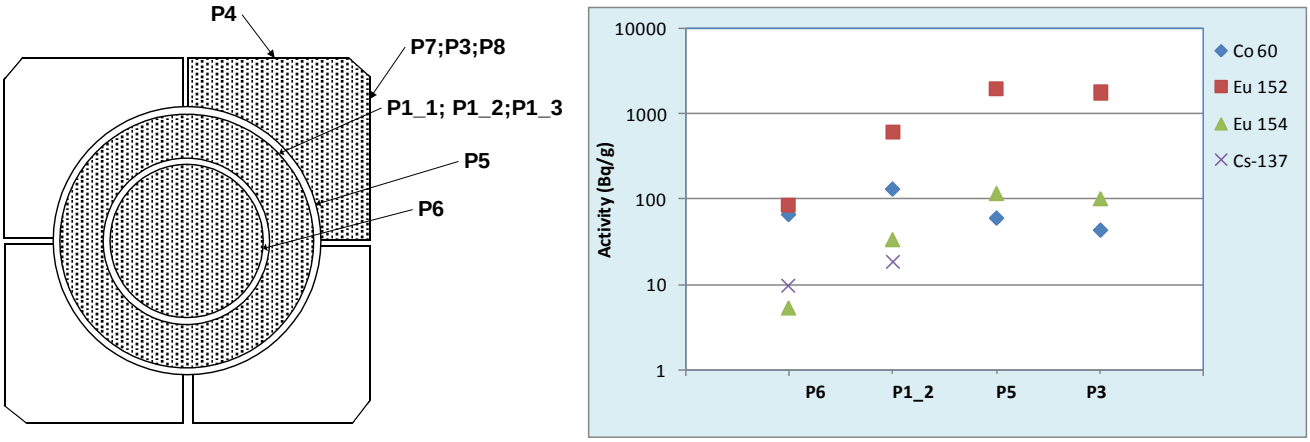

Figure 3.Activity measured values in various samples of a graphite block

Eu-152 and Eu-154 activity

Due to the presence of **Eu** as impurity (2 ppb) (*case c*) the activities of Eu-152 and Eu-154 isotopes show (Table 2 –column 4) a large increase (4 to 5 orders of magnitude) after 10 years irradiation by comparison to *case b)* – without **Eu** as initial impurity.

After 10 years of cooling (Table 3) the computed activities of Eu-152 and Eu-154 are 7.812 Bq/g and 8.216 Bq/g, respectively.

Analyzing the measured values of Eu activities in various samples (Figure 3) one could observe again a large spread of the values depending on the sample positions. The computed values seem to be close to the measured values in the sample P6.

On the other hand, the amount of the Eu impurities (2 ppb – evaluated from some previous measurements) considered in the simulations could be too small.

Cs-137 activity

The computed activity of the Cs-137 produced after 10 years irradiation is of *87.46Bq/g* and shows an increased value (*155 Bq/g*) after 10 years of cooling due to the decay of *Xe-137*. It could be observed (Figure 3) that Cs-137 has been found (measured) only in some samples; this could be due to the non-uniform distribution of the initial Th impurities.

H-3 activity

It should be noted the presence of H-3 which activity after 10 years of cooling is around 92.23 Bq/g.

3.3 Hazard parameter values

From both waste management and radiological protection point of view it could be of interest to present the activities of the first 20 dominant isotopes after 10 years of cooling (for case *c)* containing all the impurities), their associated inhalation and ingestion doses (Table 4), as well as the clearance indexes (Table 5).

It should be mentioned that the ingestion and the inhalation doses shown in the tables represent the potential radiological hazards that estimate the effect on humans of the ingestion or inhalation of radionuclides.

Table 4. Activities and doses of the main 20 dominant nuclides

	Nuclide	Activity (Bq/kg)		Nuclide	Ingestion (Sv)		Nuclide	Inhalation (Sv)
	Total	**4.706E+07**		**Total**	**4.432E-02**		**Total**	**2.060E+00**
1	**Ar 39**	4.214E+06	1	**Ar 39**	1.011E-02	1	**Ar 39**	1.011E+00
2	**La140**	6.094E+05	2	**Sr 90**	5.404E-03	2	**Ac227**	5.260E-01
3	**Sr 89**	5.326E+05	3	**I 131**	3.602E-03	3	**Pa231**	3.552E-01
4	**Rb 89**	5.324E+05	4	**Ce144**	2.285E-03	4	**Sr 90**	3.088E-02
5	**Ba140**	5.291E+05	5	**Cs137**	2.016E-03	5	**Ce144**	2.329E-02
6	**Cs140**	5.223E+05	6	**Pa231**	1.801E-03	6	**Ra223**	2.099E-02
7	**Kr 89**	5.138E+05	7	**I 133**	1.447E-03	7	**Kr 85**	1.672E-02
8	**Y 91**	5.106E+05	8	**Sr 89**	1.385E-03	8	**Th232**	1.026E-02
9	**Sr 91**	5.101E+05	9	**Ba140**	1.376E-03	9	**Th227**	9.395E-03
10	**Ba139**	5.074E+05	10	**Y 91**	1.225E-03	10	**Th228**	6.328E-03
11	**Ce141**	5.069E+05	11	**La140**	1.219E-03	11	**Cs137**	6.049E-03
12	**La141**	5.068E+05	12	**Ac227**	1.052E-03	12	**Y 91**	4.544E-03
13	**Ba141**	5.066E+05	13	**Te132**	9.011E-04	13	**Sr 89**	4.207E-03
14	**Cs139**	5.065E+05	14	**Zr 97**	6.150E-04	14	**U 233**	3.577E-03
15	**Rb 91**	5.019E+05	15	**Y 93**	5.530E-04	15	**Ru106**	3.300E-03
16	**Kr 90**	4.883E+05	16	**Kr 88**	5.395E-04	16	**Ba140**	3.069E-03
17	**Y 92**	4.873E+05	17	**Pr143**	5.327E-04	17	**U 232**	2.801E-03
18	**Sr 92**	4.870E+05	18	**Y 90**	5.213E-04	18	**Zr 95**	2.475E-03
19	**Cs141**	4.753E+05	19	**Ce143**	4.883E-04	19	**Ce141**	1.926E-03
20	**Cs138**	4.655E+05	20	**Xe133**	4.375E-04	20	**Ra228**	1.363E-03
Rest		3.315E+07		Rest	6.810E-03		Rest	1.620E-02

As described in the FISPACT code manual the doses are evaluated taking into account the coefficients termed as "committed effective doses per unit intake" " - *e-ing* for ingestion and *e-inh* for inhalation, respectively. The coefficients are used to convert the activity (Bq) of an ingested/inhaled radionuclide into the dose (Sv); for occupational exposure, all exposed persons are adults and therefore the period of time over which the committed effective dose is assessed is 50 years, irrespective of the age at intake.

They could have different values for the same radionuclide being estimated taken into account additional aspects as the fraction of the intake that remains in the body which depends also on the chemical and physical form and the route of the intake.

As specified by the authors of the FISPACT, the code libraries contain the maximum values of these coefficients; thus, from this point of view the dose values provided by FISPACT code could be overestimated.

Table 5. Clearence index values

	Nuclide	**Clearence Index**
	Total	**4.62E+04**
1	**Kr 89**	1.66E+03
2	**Cs137**	1.55E+03
3	**Br 86**	1.51E+03
4	**Br 88**	1.50E+03
5	**Br 87**	1.42E+03
6	**Rb 91**	1.25E+03
7	**Rb 89**	1.24E+03
8	**La142**	1.09E+03
9	**Rb 90**	1.05E+03
10	**La144**	1.05E+03
11	**Se 85**	1.02E+03
12	**Cs140**	9.33E+02
13	**Rb 94**	9.07E+02
14	**Sr 93**	8.48E+02
15	**Kr 88**	8.30E+02
16	**Rb 93**	7.14E+02
17	**As 84**	6.74E+02
18	**Kr 90**	6.69E+02
19	**I 136**	6.44E+02
20	**Cs141**	6.17E+02
Rest		2.50E+04

As could be observed the total clearance level is very large after 10 years of cooling and still remains high even after hundreds of years due to long-lived isotopes.

4 Conclusions

The analysis performed leads to the conclusion that the graphite undergoing long term irradiation cannot be released from regulatory control even after a long cooling time.
The total activity and total clearance index values are due to the long lived isotopes produced by the graphite impurities during irradiation.
It should be also noted that the initial weight of Eu as impurity in the graphite seems to be higher than the amount (2 ppb) taken into consideration in the present calculations.
It is also important to emphasize that the impurities could have non-uniform distributions in the graphite volume (even in the same graphite block along a distance of few centimeters the impurity weights are different as provided by the measurements performed).

5 Bibliography

1. **Roth, Csaba.** *Dispozitiv coloana termica.* IRNE. 1986. Project. 660-0.

2. **MCNP4C.** *Monte carlo N-Parcticle Transport Code System.* ORNL. 2000. CCC700.

3. **Roth, Csaba.** *Adaptare coloana termica.* SCN. 1999. Project. 2-2-18-7-0.

4. **Roth, Cs.; Dobrea D; Aioanei, L; Gugiu, D; Garlea, I; Keleman, C; Garlea, C.** *Dezvoltarea de etaloane si facilitati de iradiere in spectre neutronice de referinta.* SCN. 2004, RI 6995.

5. **R. A. Forrest**, The European Activation System: EASY – 2005, UKEA FUS 513; FISPACT -2005 User's Manual, UKEA FUS 614.

.

6 List of Figures

7 List of Tables