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Chemical removal of Co-60, Eu-152 and Eu-154 from the graphite irradiated in TRIGA reactor

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Document title

Chemical removal of Co-60, Eu-152 and Eu-154 from the graphite irradiated in TRIGA reactor

Executive summary

In WP 4.3, experiments on the chemical extraction of Co-60 and Eu-152 from irradiated graphite have been successfully performed using different acids at various concentrations. The better efficiency in Co-60 removal was obtained for HCl 50M, while H_2SO_4 was more efficient in Eu-152 removal.

Revisions

Rev.	Date	Short description	Authors	Internal Review	Task Leader	WP Leader
01	01/06/2010	Issue	Carmen Arsene Marius Iordache INR	Daniela Diaconu INR	Carmen Arsene INR *Signature*	Werner von Lensa FZJ *Signature*

The irradiated graphite from the thermal columns of TRIGA reactor contains important quantities of Eu-152 as well as Eu-154, Co-60, Cs-137, C-14 and H-3. In order to minimize the waste volume and inventory for their final disposal, INR develops and optimize methods for the chemical decontamination of the irradiated graphite based on strong solutions. The influence of the process parameters on the decontamination factor is investigated.

1. Milestones

I-graphite samples used in experiments - radionuclide activity characterization

According to the analyses performed by gamma ray spectrometry, respectively gamma spectrometry and the analyses performed by proportional counter, the irradiated graphite from TRIGA reactor (referring at the samples used in experiments) contains/is characterized by the following activities:

Co-60: 52.35 ± 2 Bq/g
Eu-152: 1333.20 ± 24 Bq/g
Eu-154: 67.85 ± 4 Bq/g
Cs-137: 3.30 ± 1Bq/g

C-14: 7.2± 1.5 Bq/g
H-3: 142.8± 1.5 Bq/g

Bglobal 328.83 Bq/g

2. Leaching tests of i-graphite

In view of getting some information with regards the behavior of radionuclides in contact with water a certainly quantity of i-graphite was contacted for 3, 6 and respectively 100 days with demineralized water. The leached beta total activity and the leached radionuclides are presented in the table 3.2.1.

Table 2.1 The leached activity during the water contact

No. sample	Contacting time (days)	% leached beta global activity*	% leached activities			
			Co-60	Cs-137	Eu-152	Eu-154
a-g	100	52.12	9.13	100.00	13.43	8.14
b-g	3	15,13	2.50	-	11.22	6.82
c-g	6	22,41	1.09	22.05	12.11	-

3. Experimental methods for activity removal

The experiments for removal of Co-60, Eu-152 & Eu-154 consisted in contacting the i-graphite (under the powder form) with different acids and mixtures of acids.

Table 3.1 The experiments performed for radionuclide chemical removal

No. exp.	Removal solution	No. Exp.	Removal solution
1-g	HNO_3 65%	9-g	HNO_3 65% : H_3PO_4 85% (1:1)
2-g	HNO_3 65%: demi water (1:1)	10-g	(HNO_3 65% + H_3PO_4 85%=1:1):demi water (1:1)
3-g	HCl 37%	12-g	(HCl 37%+ HNO_3 65%=1:1): demi water (1:1)
4-g	HCl 37%: demi water (1:1)	13-g	HCl 37%:H_2SO_4 98% (1:1)
5-g	H_2SO_4 98%	14-g	(HCl 37%: H_2SO_4 98%=1:1): demi water (1:1)
6-g	H_2SO_4 98% : demi water (1:1)	15-g	oxalic acid 10%
7-g	H_3PO_4 85%	16-g	citric acid 10%
8-g	H_3PO_4 85%: demi water (1:1)	17-g	oxalic acid 10% : citric acid 10%

For all experiments the contacting time was 6 days. The vials were periodically mechanically shaken. After the contacting period a known quantity of demineralized water was added and then they were centrifuged for 7 minutes and filtrated. The both, filtrates and graphite samples, were measured.

■ *Measuring the graphite*

Known quantites of wet graphite were add on the experimental plates and the layer was uniformized by adding 2-3 drops of etilic alcohol. The samples were dried under the UV lamp and an incrustation was given (protecting layer of toluene+polistiren). The samples thus prepared were measured.

Table 3.2 The beta global activity of the graphite after chemical treatment and the removal efficiency

No. exp.	Wet graphite sample (g)	Dry graphite in sample* (g)	Beta global activity of dried graphite after decontamination (Bg/g)	Removal efficiency** (%)
1-g	0.75	0,38	243,01	26,10
2-g	0.72	0,39	207,80	36,81
3-g	0.68	0,36	197,71	39,87
4-g	0.68	0,34	242,99	26,11
5-g	0.74	0,29	125,39	61,87
6-g	0.78	0,31	133,06	59,53
7-g	0.74	0,34	119,17	63,76
8-g	0.71	0,32	145,11	55,87
9-g	0.71	0,28	227,08	30,94
10-g	0.79	0,38	229,81	30,11
12-g	0.73	0,37	244,59	25,62
13-g	0.79	0,36	183,65	44,15
14-g	0.74	0,45	95,75	70,88
15-g	0.71	0,44	241,59	26,53
16-g	0.74	0,42	257,53	21,68
17-g	0.73	0,43	282,10	14,21

g = graphite, P1- g, P2 -g = sample i-graphite powder

the samples P1-g, P2-g, b-g, c-g, 1-4g, 9-13g were measured in infinite thick source
the samples 5-8g, 13-g and 14-g were measured in infinite thin source
* the quantity of dried graphite was calculated by referring the quantity of wet graphite taken as sample at the quantities of initial dry and wet graphite (weight)
** it has been referred to the average of i-graphite activity (1g = 328.83 Bq)

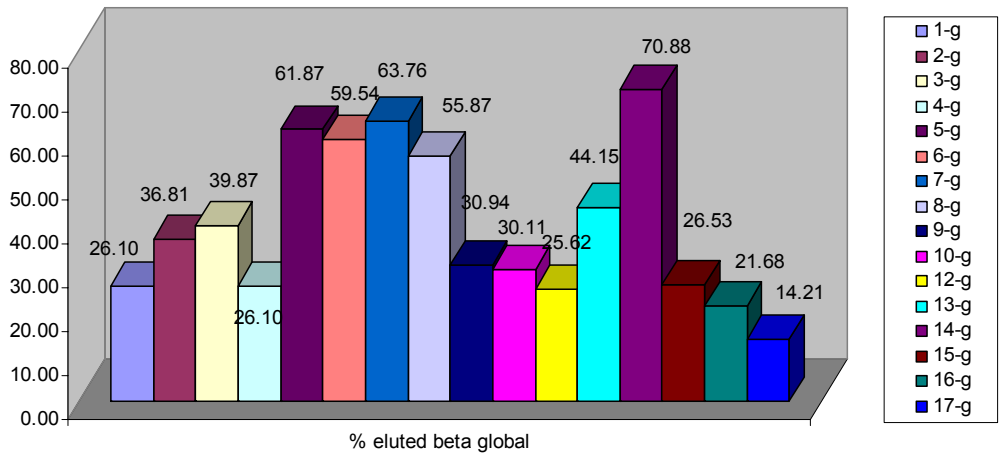

Fig. 3.1 Beta total removal efficiency

It is shown within the set of experiments the decontamination efficiency with regards at the global beta activity is the highest (71%) in case of sample no. 14 (decontamination solution: 2.5 ml HCl 37%+ 2.5 ml H_2SO_4 98%+ 5 ml demineralized water), followed by the samples 5, 6, 7, 8 (efficiency = 59-63%) – the experiments where H_2SO_4 98% and H_3PO_4 85% were used (experiments no. 5 and 7) and in mixture with demineralized water, in volumetric ratio 1:1 in the experiments 6 and 8.

Table 3.3 Decontamination tests results - Co-60, Eu-152 & Eu-154 removal

No. exp.	Activity (Bq/g)				% radionuclides removed			
	Co-60	Cs-137	Eu-152	Eu-154	Eu-154	Cs-137	Eu-152	Eu-154
1-g	21.52±3	4.53±3	932.65±66	50.28±6	58.89	-	30.03	25.90
2-g	22.51±3	2.81±1.4	930±63	48.56±5	57.00	15.11	30.23	28.43
3-g	9.47±2	-	900.13±60	49.43±6	81.91	100	32.47	27.15
4-g	19.02±3	8.27±4	9.6.05±65	51.29±6	63.67	-	92.79	24.41
5-g	6.2±2	-	372.94±28	20.29±3	88.16	100	72.02	70.10
6-g	5.16±1.5	-	392.78±28	20.73±3	90.14	100	70.53	69.45
7-g	8.85±2	-	526.60±38	30.65±4	83.09	100	60.50	54.83
8-g	9.17±2	-	520.36±38	28.88±4	82.48	100	60.96	57.44
9-g	7.19±2	-	479.09±33	27.68±3	86.27	100	64.06	59.20
10-g	13.38±3	-	576.81±42	32.01±4	74.44	100	56.73	52.82
12-g	17.43±2	-	863.53±62	46.98±5	66.70	100	35.22	30.76
13-g	6.43±2	3.63±2.1	551.81±40	29.89±4	87.72	-	58.60	55.95
14-g	7.95±2	-	591.87±41	33.19±4	84.81	100	55.60	51.08
15-g	17±3	4.11±3	1009.13±70	58.65±6	67.53	-	24.30	13.56
16-g	31.34±3	-	878.47±23	46.13±4	40.13	100	34.10	32.01
17-g	30.32±4	-	1148.51±82	63.9±7	42.08	100	13.84	5.82

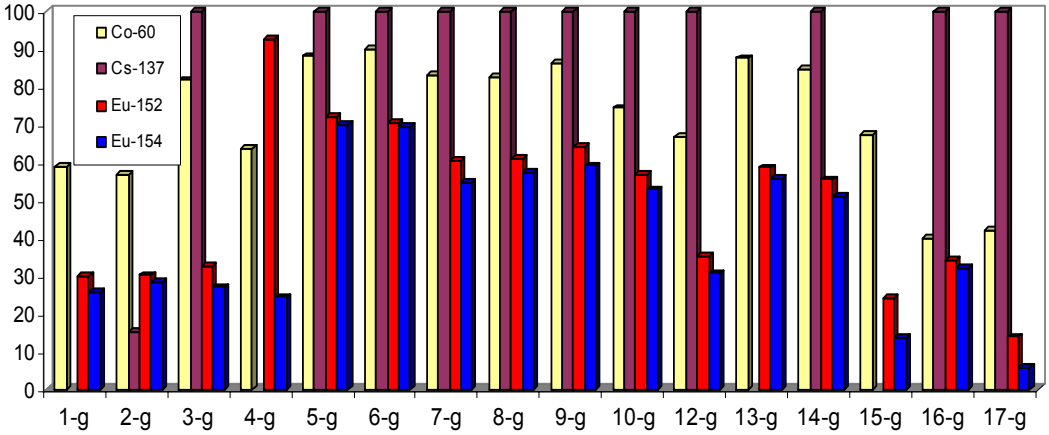

Fig. 3.2 Co-60, Eu-152 & Eu-154 removal efficiency

This diagram shows the efficiency is maximum taking into consideration all radionuclides for exp. 5 and 6 (meaning decontamination with H_2SO_4 solution).
Even if it is observed a procent of 100% efficiency for Cs-137 we can not admit it because it was initially in a very low activity (3.3 Bq/g) such as it is normal to get such values (also it is well known the high leaching rate of caesium).

In view of determining the content of C-14 and H-3, Perkin Elmer sample Oxidizer model 307, was used where the sample is combusted and the two radionuclides are separated by the others. The samples were then analyzed by beta spectrometry measurement in Tri-Carb Packard 2100.

Table 3.4. Decontamination tests results - Co-60, Eu-152 & Eu-154 removal

Nr. exp	g sample	Bq/g
PrB-1	0.29	<0.23
PrB-2	0.32	7.18
		142.8 (H-3)
a-g	-	-
b-g	0.43	<0.23
c-g	0.32	<0.23
1-g	0.31	<0.23
2-g	0.30	<0.23
3-g	0.30	<0.23
4-g	0.31	<0.23
5-g	0.29	<0.23

Nr. exp	g sample	Bq/g
6-g	0.31	<0.23
7-g	0.31	<0.23
8-g	0.30	<0.23
9-g	0.37	<0.23
10-g	0.27	<0.23
12-g	0.30	<0.23
13-g	0.35	<0.23
14-g	0.36	<0.23
15-g	0.35	<0.23
16-g	0.35	<0.23
17-g	0.30	<0.23

4. Conclusion

Beta global activity efficiency:

71% - using decontamination solution: 2.5 ml HCl 37%+ 2.5 ml H_2SO_4 98%+ 5 ml demi water
59-63% - using decontamination solution: H_2SO_4 98%, H_3PO_4 85% and diluted.

Co-60, Eu-152 & Eu-154 removal efficiency:

per g i-graphite	Co-60	Eu-152	Eu-154
3.3 ml H_2SO_4 98%	88 %	72%	70%
6.7 ml H_2SO_4 ~ 50%	90%	70%	69%

5. Perspectives

Deliverables - Plan for the proposed work of the next 12 months

- Continuing the experiments for i-graphite decontamination with acids or mixtures of acids (different contacting time, different concentrations, different ratios), determination of decontamination factor in view of establishing the optimum agent.
- trapping CO2 during decontamination and measurement - in a solution (ex.NaOH solution) - and to design a method for capturing it during experiments