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First Characterisation Report of HFR EU1bis irradiation experiment samples

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Abstract

Considering first the low enriched uranium oxide kernels, irradiated samples have been identified (pebble piece, particles), coming from the HFR EU1bis irradiation experiment. These samples have been first transported from Petten to ITU using a TN106 cask, taking benefit from another transport in the frame of the RAPHAEL project. The transport between ITU and the Atalante facility has been carried out with a RD15IIB cask when the licence was available. Then, the characterisation programme started with very first examinations before going on with non destructive and destructive analysis. This report presents the first characterisation results obtained in Atalante facility.

Revisions

Rev.	Date	Short description	Author	WP Leader	SP Leader	Coordinator
01	1/04/10	Final issue		P. Bros CEA	W. Von Lensa FZJ	E. Bogush AREVA NP

CONTENTS

1 Introduction

The generation of waste from the operation of nuclear power plants, and specifically the VHTR one is an issue of major importance with regard to sustainable development, and receives much more attention than in the past when the main emphasis was on the technological development of the reactor systems and high conversion fuel cycles. In this field, an ambitious characterisation programme is scheduled in Atalante facility with HTR samples.

The objective of the RAPHAEL sub project on the back end of the fuel cycle is to study the characteristics and performance of HTR fuel, in particular, the behaviour in direct geological disposal conditions. Existing characterization data from past investigations, in France, Germany and UK, have to be completed. In this field, HTR samples have been transported to Atalante facility in order to start the characterisation programme. This programme includes :

- Non destructive examinations,
- Destructive examinations on aqueous samples coming from chemical preparations on irradiated fuel pieces and solid samples (microstructure analysis).

This report presents, first, the received samples, and, the first characterisation results.

2 RAPHAEL Project Samples

It has been decided to use HFR EU1BIS samples. Thus, they have been transported in a previous transport between HFR reactor and ITU, using the TN106 cask. The transport between ITU and the Atalante facility has been performed with a RD15IIB cask under a “A” type licence obtained on September, 2007.

2.1 Samples characteristics

The samples have been burned in the HFR reactor. Table 1 gives the main characteristics of the pebbles and coated particles. Only a part of these samples have been received in the Atalante facility. In details, there were :

i. A graphite half shell without fuel,
ii. Two small sticks each about 10 coated particles,
iii. A small piece of the GLE4.2 pebble.

Total material masses are the following:

- Before irradiation
  - Uranium, 73 mg
  - ^{235}U, 12 mg.
- After irradiation
  - Uranium, 60 mg
  - ^{235}U, 3,34 mg
  - Plutonium, 2,35 mg

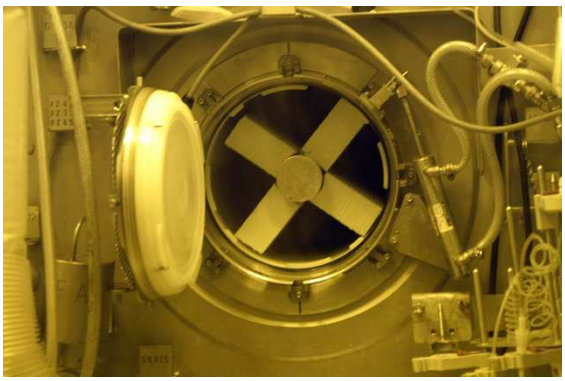
2.2 Main data

The irradiation experiment started on September, 2004 for ending on October, 2005. The maximum burnup was equal to 15,5 % FIMA. The transport has been carried out on December. Considering about two years of cooling time, table 2 indicates the main isotopes activities, based on calculations performed with data given by Klaas Bakker (HFR) and Ludovic Mariette (TNI).

2.3 Material transportation

HTR samples have been received on December, the 14th, in a hot cell devoted to analysis (see photos).

Figure 1 : a) cask arrival b) connexion to the hot cell



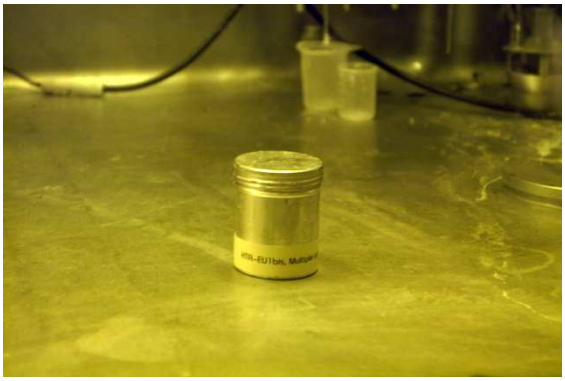


Figure 2 : a) hot cell introduction b) container in the cell

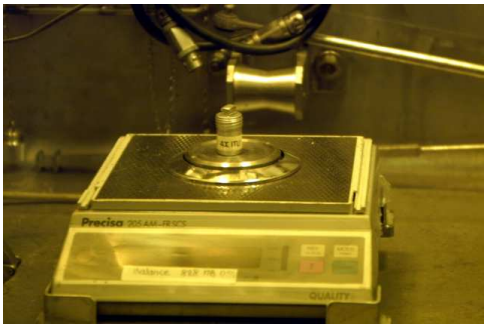


Figure 3 : a) samples packaging b) weighting control

3 First Characterisation results

In WP1, the non destructive analyses in hot cell have been achieved on the three HTR fuel samples of irradiated coated-articles from EU1bis irradiation experiment. Optical microscope specially implemented to hot cell examinations and gamma spectrometry measurements have been chosen at this step of the characterisation process. In an investigating approach, some data have also been monitored on the photon spectrum of K-edge X rays fluorescence emitted by the sample submitted to an high level X radiation in order to detect the presence of uranium and plutonium.

- ***Graphite half shell***

The first sample made of a graphite piece (mass 0,655 g) present a low dose rate (0,5 mGy/h). Its gamma spectrum is focussed on ^{137}Cs radioisotope emission (78%), the minor emitters are ^{134}Cs, ^{241}Am et ^{234}Am. None ^{60}Co, ^{106}Ru as U and Pu have been detected.

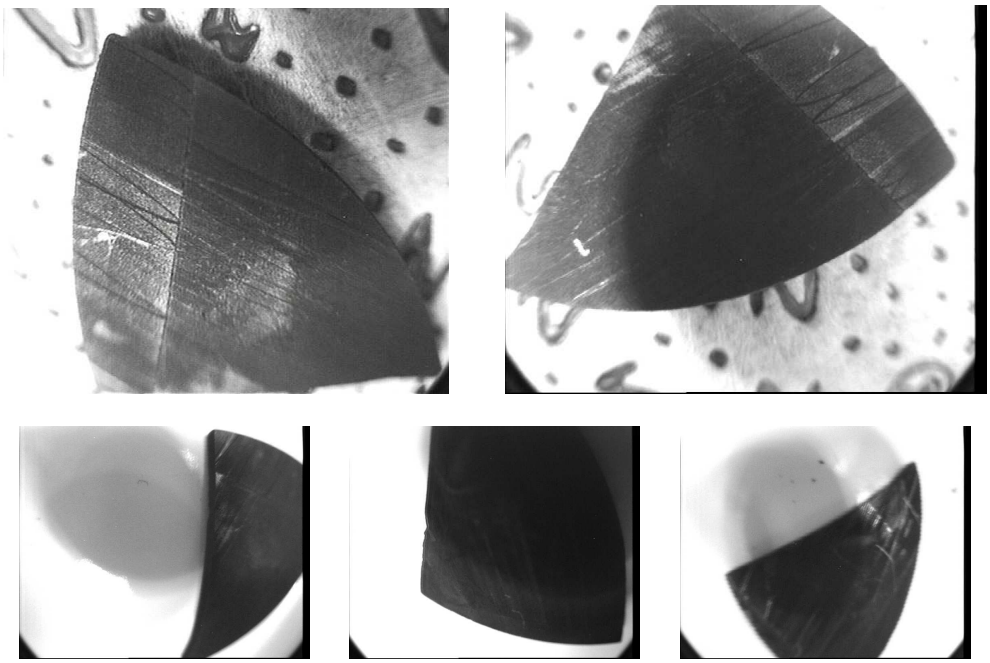


Figure 4 : Graphite half shell photos

- ***Two small sticks***

The second sample (mass 0,616 g) consisted in two sticks of about 2 mm of diameter and 20 mm long. Its higher dose rate level around 20 mGy/h indicated more gamma emitters' abundance. Its gamma spectrum shows the presence of ^{137}Cs (34%), ^{106}(Ru+Rh) (31%), ^{144}(Ce+Pr) (21%), ^{134}Cs (11%). The emission of ^{241}Am, ^{243}Am, ^{125}Sb, ^{154}Eu, ^{155}Eu have been detected but remains of minor importance (less than 3 %). During their handling in the shielded cell, the sticks have been partially reduced in small pieces and powder because of their brittle behaviour (cf. photos in the next paragraph). Due to the physical state of these samples, the K-edge measurement for U and Pu couldn't have be done.

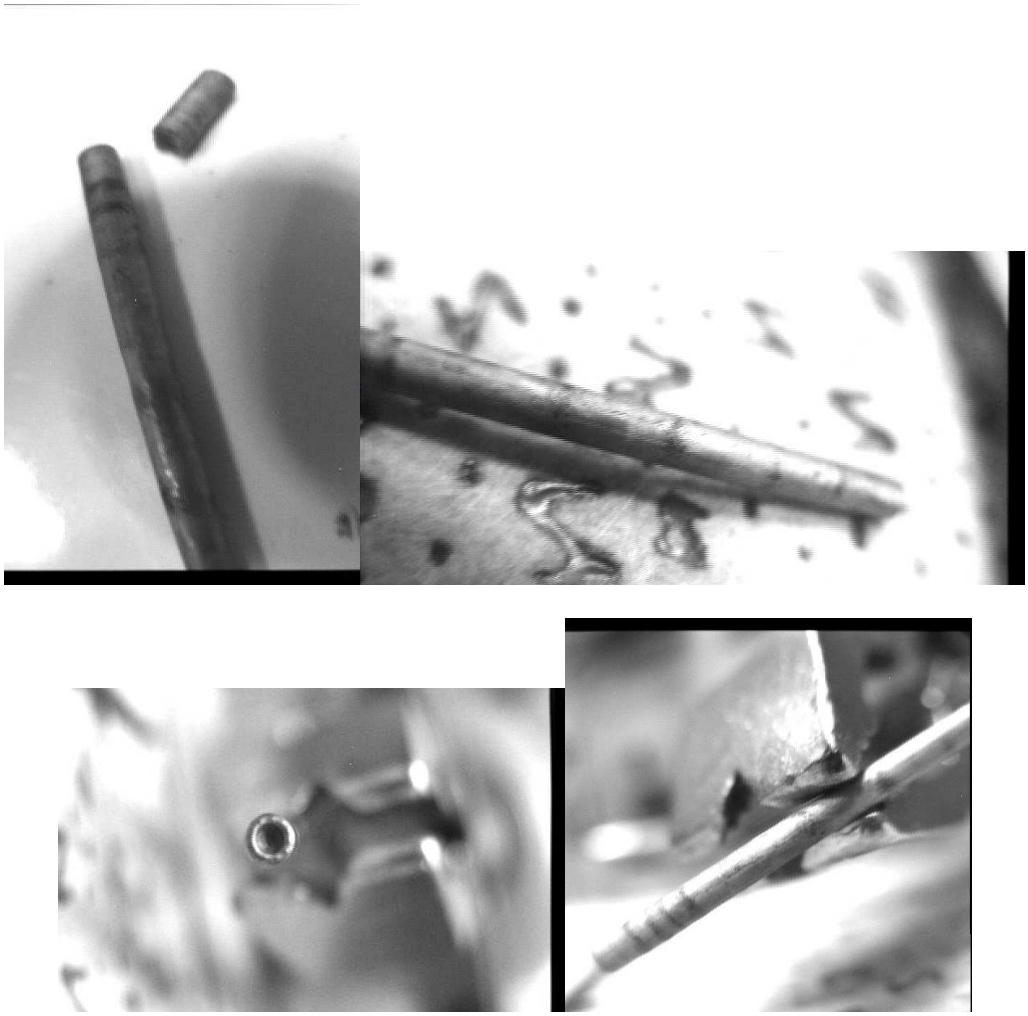


Figure 5 : sticks photos

- ***Small piece of the GLE4.2 pebble***

The third sample type was constituted by one massive cylinder (10 mm diameter – 12 mm heigh – mass 1,363 g) of graphite containing about 100 small irradiated UO_2 particles. The strong gamma emission (150 mGy/h of dose rate) represented almost the same gamma-emission spectrum as observed in the sticks case. The K-edge measurements indicated clearly the presence of U and Pu with an estimated ratio Pu/(U+Pu) close to 1,3%.

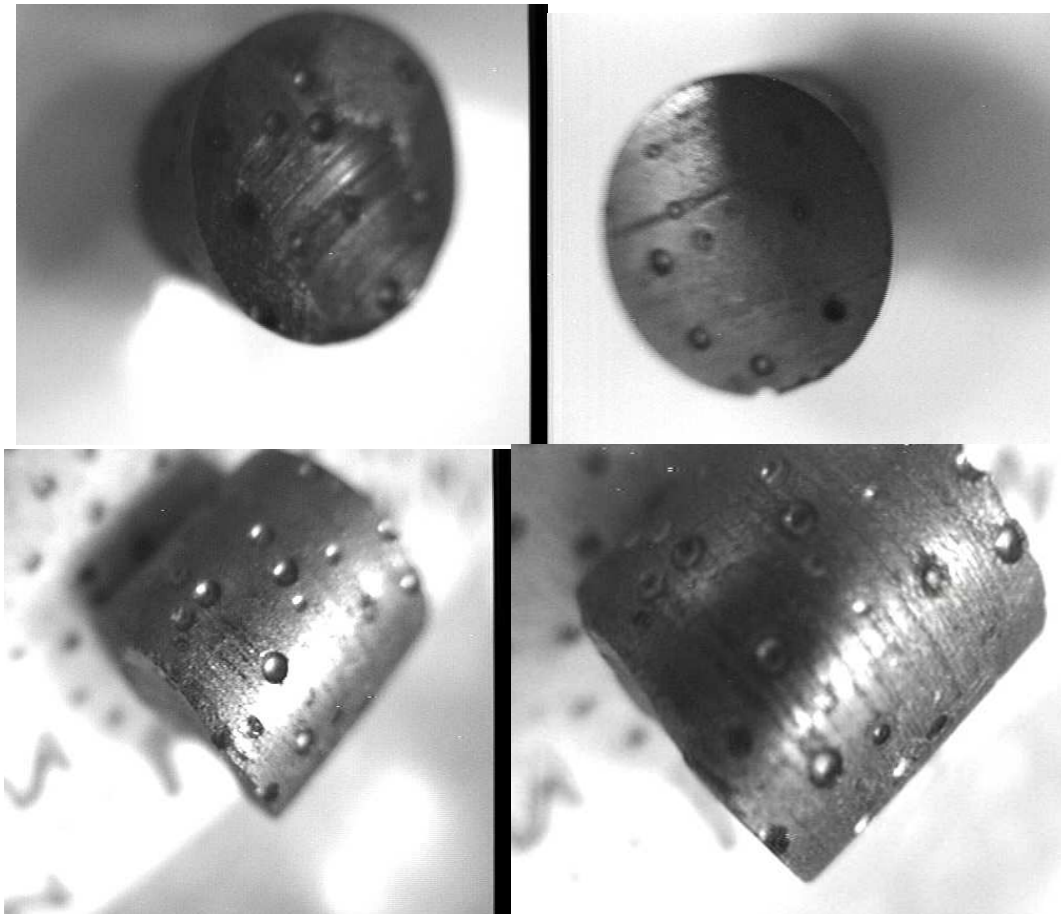


Figure 6 : GLE 4.2 pebbles piece photos

- **Prospects for destructive characterization**

After the step of sampling preparations for further micro-structural analyses, it will be possible to get back UO_2 particles from the different pieces using a mild milling. The specific solutions obtained after HTR fuel particles nitric dissolution will be analysed with techniques available in Atalante as L-edge X-ray fluorescence, ICP/AES, ICP/QMS and TIMS to determine the concentrations of actinides and some fission products.

4 Samples partition for future works

Three HTR fuel samples of irradiated coated-articles from EU1bis irradiation experiment have been separated in to two batches on view of the different destructive analysis planned. The parts created have equivalent mass. The first sample made of a graphite piece has been cut as showed in figure 7 (masses 0.263 g and 0.365 g). The second sample (mass 0,616 g) consisted in several piece of sticks of about 2 mm of diameter and CP, as visualized in figure 8. The parts have similar composition of sticks and particles (masses 0.267g and 0.297g).

The third sample type was constituted by one massive cylinder (10 mm diameter – 12 mm height – mass 1,363 g) coming from a pebble and with about 100 small irradiated UO_2 particles. It has been cut in three slices, with different thickness (masses 0.694 g, 0.231g and 0.169 g).

Some of these samples, sticks, coated particles and slice of pebble, will be now embedded and polished according to the last protocol established on old particles coming from past SILOE reactor irradiation.

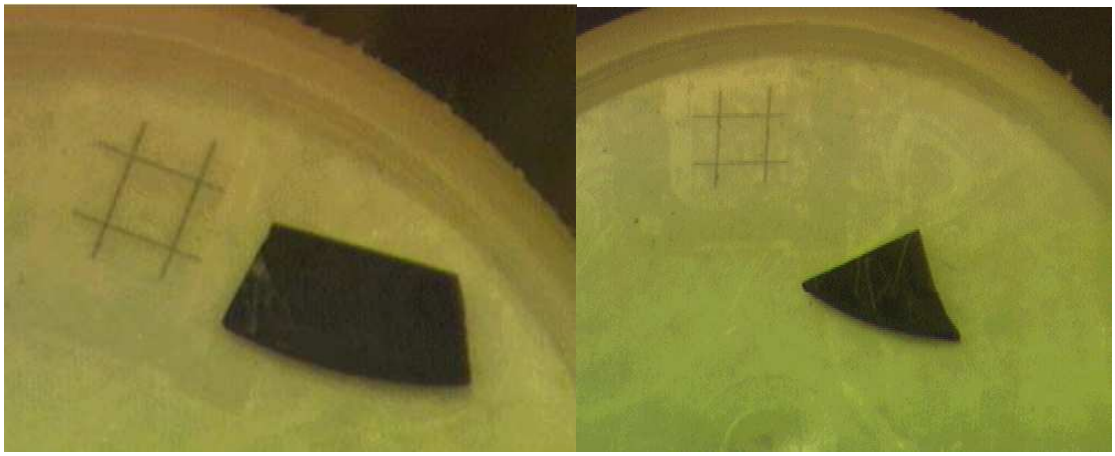


Figure 7 : Photography of graphite irradiated samples after cutting

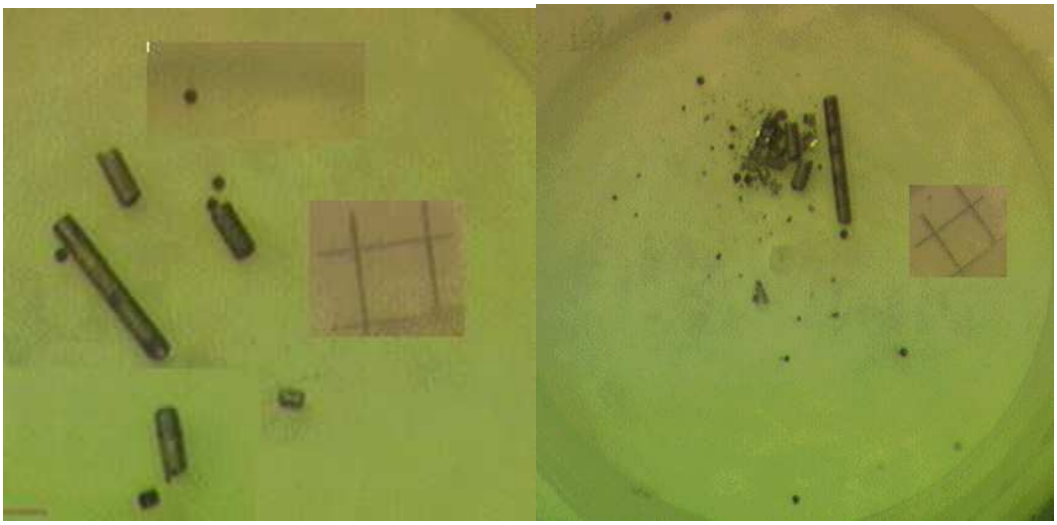


Figure 8 : Image of sticks and coated particles from second irradiated sample.

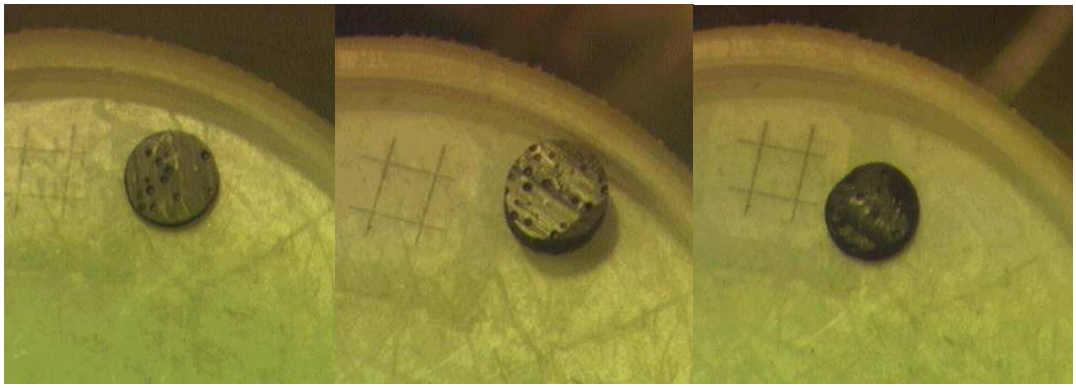


Figure 9 : Image of irradiated pebble slices. The CP locations (holes) are clearly visible

5 Conclusions

The characterisation programme will go on with non destructive et destructive analysis. It is scheduled on april, 2010, to dissolve HTR samples et to analyse the obtained solution (both chemistry and radiochemistry analysis and isotopic compositions). In parallel, microprobe and SEM characterisations will be performed on solid samples. The results and the operating protocols will be described in the final report corresponding to deliverable n°5.

6 References

- M. Fütterer, E. Toscano, K. Bakker, Irradiation results of AVR Pebbles at increased temperature and burn up in the HFR Petten, HTR 2006, Sandton, South Africa, 2006.
- Mail from K. Bakker (01/02/2007) concerning the HTR samples transport.
- C. Sciolla, Report on calculations bout the HFR EU1 bis irradiation experiment, januar 2007.
- Mail from L. Mariette (TNI) (04/05/2007).
- Mail from K. Bakker (02/02/2006) on nuclides inventory after irradiation with various cooling time.

Table 1 : Main characteristics

Particles	kernel	UO_2
	Kernel diameter (µm)	502
	Enrichment (% ^{235}U)	16,76
	Coating thickness (µm) : ▪ Buffer ▪ Inner PyC ▪ SiC ▪ Outer PyC	 92 40 35 40
	Particle diameter (µm)	916
Pebbles	U (g/pebble)	6
	^{235}U (g/pebble)	1
	Particules number	9560

Table 2 : Main isotopes activities

Isotopes	Activities after 2 years cooling time	A2 (Bq)	Ai/A2
U 233	3,54E-02	6,00E+09	5,90E-12
U 234	4,47E+01	6,00E+09	7,45E-09
U 236	5,19E+03	6,00E+09	8,65E-07
Np237	3,37E+03	2,00E+09	1,69E-06
Pu236	7,07E+02	3,00E+09	2,36E-07
Pu237	3,20E-02	2,00E+13	1,60E-15
Pu238	1,66E+07	1,00E+09	1,66E-02
Pu239	4,86E+06	1,00E+09	4,86E-03
Pu240	6,51E+06	1,00E+09	6,51E-03
Pu241	2,39E+09	6,00E+10	3,98E-02
Pu242	3,19E+04	1,00E+09	3,19E-05
Pu244	9,87E-04	1,00E+09	9,87E-13
Am241	6,29E+05	1,00E+09	6,29E-04
Am243	1,66E+05	1,00E+09	1,66E-04
Cm242	1,24E+07	1,00E+10	1,24E-03
Cm243	4,25E+04	1,00E+09	4,25E-05
Cm244	1,12E+07	2,00E+09	5,61E-03
Cm245	6,60E+02	9,00E+08	7,33E-07
Cm246	9,62E+01	9,00E+08	1,07E-07
Cm247	1,29E-04	1,00E+09	1,29E-13
Cm248	2,03E-04	3,00E+08	6,76E-13
Bk249	4,10E-01	3,00E+11	1,37E-12
Cf249	3,70E-05	8,00E+12	4,62E-18
Cf250	1,03E-02	2,00E+09	5,17E-12
Cf251	4,37E-05	7,00E+08	6,24E-14
Cf252	7,20E-03	3,00E+09	2,40E-12
KR85	7,52E+07	1,00E+13	7,52E-06
Sr89	7,95E+05	6,00E+11	1,32E-06
Sr90	4,90E+08	3,00E+11	1,63E-03
Y90	4,90E+08	3,00E+11	1,63E-03
Zr95	1,23E+07	8,00E+11	1,54E-05
Nb95	2,73E+07	1,00E+12	2,73E-05
Ru103	9,25E+04	2,00E+12	4,62E-08
Ru106	1,86E+09	2,00E+11	9,32E-03
Rh103m	9,11E+04	4,00E+13	2,28E-09
Sb125	8,77E+07	1,00E+12	8,77E-05
Te125m	2,20E+07	9,00E+11	2,44E-05
Te129	1,80E+02	6,00E+11	3,00E-10
Te129m	2,61E+02	4,00E+11	6,52E-10
Cs134	6,33E+08	7,00E+11	9,04E-04
Cs137	7,95E+08	6,00E+11	1,32E-03
Ce141	6,16E+03	6,00E+11	1,03E-08
Ce144	2,71E+09	2,00E+11	1,36E-02
Pm147	1,23E+09	2,00E+12	6,13E-04
Pm148	7,60E+01	7,00E+11	1,09E-10
Eu154	7,05E+07	6,00E+11	1,18E-04
Eu155	2,16E+07	3,00E+12	7,19E-06
		TOTAL	0,1