

## DETERMINATION OF $^{241}\text{Pu}$ BY LOW LEVEL $\beta$ -PROPORTIONAL COUNTING. APPLICATION TO CHERNOBYL FALLOUT SAMPLES AND COMPARISON WITH THE $^{241}\text{Am}$ BUILD-UP METHOD

G. ROSNER, H. HÖTZL, R. WINKLER

*GSF-Forschungszentrum für Umwelt und Gesundheit, Institut für Strahlenschutz,  
D-8042 Neuherberg (Germany)*

(Received January 2, 1992)

A chemical separation procedure is described which allows the direct determination of low  $^{241}\text{Pu}$  activities in environmental samples with a windowless gas-flow proportional counter. While current separation schemes based on anion exchange yield counting sources of sufficient purity for subsequent  $\alpha$ -spectrometry, for  $\beta$ -counting of  $^{241}\text{Pu}$  additional purification steps are required. A combination of anion exchange from 9 mol/l HCl,  $\text{LaF}_3$  precipitation and TTA extraction was found to be suitable even for the analysis of long-range Chernobyl fallout samples which contained interfering radionuclides with  $\beta$ -activities at least 3 to 4 orders of magnitude higher than usually encountered. No difference is detectable between the results of the present, direct procedure and the results of the conventional indirect method based on the build-up of  $^{241}\text{Am}$ . Average  $^{241}\text{Pu}/^{239+240}\text{Pu}$  ratios in air and deposition samples taken at Neuherberg near Munich were  $70 \pm 6$  with the present procedure and  $66 \pm 9$  from  $^{241}\text{Am}$  build-up.

### Introduction

Isotopes of plutonium have entered the environment as a result of nuclear weapons testing on a global scale, and from nearby testing sites, accidents and nuclear installations on a local scale. Among the isotopes of plutonium which are of interest in environmental studies (Table 1),  $^{241}\text{Pu}$  is the only  $\beta$ -emitter. Due to its low maximum  $\beta$ -energy of 20.8 keV,  $^{241}\text{Pu}$  is difficult to determine and therefore it is sometimes neglected, although in most cases it contributes most to the total plutonium radioactivity in environmental samples.  $^{241}\text{Pu}$  is the precursor of  $^{241}\text{Am}$ , an  $\alpha$ -emitter with a long residence time in biota and, therefore, it is radiologically important. Furthermore, the activity ratio of  $^{241}\text{Pu}$  to  $^{239+240}\text{Pu}$  is specific for a particular nuclear application and thus allows identification of plutonium sources and dating of deposits.

In environmental samples  $^{241}\text{Pu}$  is often determined indirectly by an  $\alpha$ -spectroscopic measurement of the daughter nuclide  $^{241}\text{Am}$ , after an ingrowth period of several years.<sup>1,2</sup> As a direct counting method, liquid scintillation counting has been reported. This technique is, however, especially suitable for samples of an intermediate  $^{241}\text{Pu}$  activity level, since the counter background is comparatively high. Typical applications of liquid

Table 1  
Nuclear data of plutonium isotopes relevant to environmental studies

Nuclide	Half-life, y	Radiation energy and abundance			
		Alpha		Beta	
		MeV	%	keV	%
$^{238}\text{Pu}$	87.7	5.499	71.0		
		5.456	28.8		
$^{239}\text{Pu}$	$2.411 \cdot 10^3$	5.156	73.0		
		5.143	15.1		
		5.105	11.7		
$^{240}\text{Pu}$	6563	5.168	72.9		
		5.124	27.0		
$^{241}\text{Pu}$	14.4	4.897	$2.0 \cdot 10^{-3}$	20.8	99.998
$^{241}\text{Am}^*$	432.7	5.486	85.2		
		5.443	12.8		

\*Daughter of  $^{241}\text{Pu}$ .

scintillation counting are, therefore, samples from regions with elevated fallout levels like Scandinavia,<sup>3</sup> samples from nuclear technology, like effluents,<sup>4-6</sup> or samples from weapons testing sites.<sup>7</sup> In these studies, lower limits of detection from 35 to 65 mBq of  $^{241}\text{Pu}$  have been reported. Mass spectrometry seems only in few cases sensitive enough for the analysis of  $^{241}\text{Pu}$  at environmental levels, and requires great instrumental efforts.<sup>8-10</sup> Few attempts have been reported on direct  $\beta$ -counting with an internal gas flow proportional counter.<sup>11,12</sup>

In environmental samples, the available activity of plutonium is in general very low, e.g., typical specific  $^{239+240}\text{Pu}$  activities from Germany are: air, average 1976-1981,  $0.4 \mu\text{Bq} \cdot \text{m}^{-3}$ ; deposition, average 1976-1981,  $0.35 \text{ mBq} \cdot \text{l}^{-1}$ ; agricultural soil, 1982,  $200 \text{ mBq} \cdot \text{kg}^{-1}$ ; biological samples, from 0.059 to  $44 \text{ mBq} \cdot \text{kg}^{-1}$ .<sup>13-15</sup> Therefore, large sample sizes are required, e.g., several tens of thousands of cubic meters of air, 10 to 100 liters of rain water, several kilograms of biological material, etc.<sup>16</sup>

The low plutonium activity available in most environmental studies makes it also desirable to determine as many nuclides as possible from one sample, since otherwise the efforts of preparing and chemical processing large samples are multiplied. One step in this direction is to use the thin counting sources required for the  $\alpha$ -spectrometric determination of plutonium isotopes also for a subsequent determination of  $^{241}\text{Pu}$  by  $\beta$ -counting.

For this purpose, a windowless gas flow proportional counter was built and calibrated with standard solutions of plutonium radionuclides. This has been described in a previous publication.<sup>17</sup> The present paper describes a suitable radiochemical separation procedure, and the application of the low level counter to the analysis of environmental field samples, namely air and deposition samples collected at Neuherberg near Munich after the Chernobyl accident. These samples contained unusually high amounts of many radionuclides, in addition to the levels of natural and global fallout radioactivity and of stable elements as well, which are usually encountered. Being contaminated by actual atmospheric processes, they could be expected to allow a very realistic test of the entire procedure. The results obtained are compared with the results of the conventional analysis by build-up of  $^{241}\text{Am}$ . Furthermore, the respective techniques themselves are compared.

## Experimental

### *Reagents*

All reagents were of standard analytical grade.  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  were used as tracer nuclides.

### *Apparatus*

For the  $\beta$ -counting of  $^{241}\text{Pu}$  an anticoincidence shielded windowless gas flow proportional counter with an effective detector diameter of 25 mm, operated with P-10 gas at atmospheric pressure, was used. This detector, which is specially designed for the counting of low energy  $\beta$ -emitters, and the associated electronics have been described by us previously.<sup>17</sup> Alpha-spectrometric measurements of the plutonium and americium isotopes were carried out in large area Frisch grid ionization chambers<sup>18</sup> or with commercial silicon surface barrier detectors. Counting times were usually 1000 min in the ionization chambers, and several 1000 minutes in the semiconductors.

### *Sampling and sample preparation*

Aerosol samples were collected at Neuherberg near Munich fortnightly at a height of 1.5 meters above ground on 0.5 m<sup>2</sup> organic fiber filters (Microsorban 99/97, Delbag; these filters are no longer available and are now replaced by Viledon FA 2311, Freudenberg) at an air flow rate of 600 m<sup>3</sup> · h<sup>-1</sup>. Total deposition was collected 1 m above ground in several tubs, each of 0.6 m<sup>2</sup> area. These samples include dry fallout, since the detector bases were kept covered with deionized water during dry periods.

75 mBq of  $^{242}\text{Pu}$  in 50  $\mu\text{l}$  of 2 mol/l  $\text{HNO}_3$  were added as a tracer. Aerosol samples were ashed in a muffle furnace at 550 °C. The deposition samples were evaporated in a large rotary evaporator (Büchi R 170). The ashes as well as the evaporation residues were then digested successively several times with 65%  $\text{HNO}_3$ , 40% HF and 70%  $\text{HClO}_4 + \text{H}_2\text{O}$  1 + 1. These procedures have been described in detail previously.<sup>16</sup>

### *Chemical processing*

Plutonium was first separated from the aerosol and deposition samples according to a procedure aimed at subsequent  $\alpha$ -spectroscopic determination, which is based on anion exchange and has been described by us previously.<sup>16</sup> However, to remove quantitatively the unusually large activities of interfering radionuclides present in the Chernobyl samples, additional purification steps proved to be necessary. An adaptation of a procedure described by KRACKE und BUNZL<sup>19</sup> was found to be suitable. After the initial anion exchange separation, the plutonium is present in 50 ml of 1.2 mol/l HCl solution containing hydrogen peroxide. To this solution, 1 ml of La carrier (20 mg  $\text{La}^{3+}/\text{ml}$ ) is added, and the solution is boiled down to 2–3 ml in a glass beaker, made up to 100 ml with deionized water and a pH of 1.5–2 is adjusted with ammonia. Subsequently plutonium is coprecipitated with  $\text{LaF}_3$  by adding 10 ml of 5% NaF solution in distilled water. The precipitate is allowed to settle for at least 1–2 hours, and then centrifuged. The residue is dissolved in a total of 40 ml of 1 mol/l  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ , and the tetravalent state of plutonium is adjusted by adding 5 drops of a freshly prepared 12%  $\text{NaNO}_2$  solution in distilled water and standing for 15 minutes. Pu is then extracted by vigorous shaking for 20 minutes with 20 ml of a 5% solution of thenoyltrifluoroacetone (TTA) in benzene. The organic phase is scrubbed twice with 20 ml deionized water each by shaking for 5 minutes. The plutonium is back extracted with 10 ml of 9 mol/l HCl, by shaking vigorously for 15 minutes. This back extraction is repeated once. The aqueous phase is purified by shaking for 1 minutes with 10 ml of diisopropyl ether. From the aqueous phase, a counting sample is prepared by electrolysis onto a stainless steel disc from a slightly acidic ammonium sulfate medium as described previously.<sup>16</sup>

The stainless steel discs with the electrodeposited plutonium were stored for about 4 years to allow build-up of  $^{241}\text{Am}$ . Then, the active deposit was dissolved by repeated leaching with 3 ml of warm 7 mol/l  $\text{HNO}_3$ . As a tracer, 50  $\mu\text{l}$  of  $^{243}\text{Am}$  solution in 2 mol/l  $\text{HNO}_3$ , containing 60 mBq  $^{243}\text{Am}$ , were added to the first portion of the leaching solution. Less than 0.5% of the deposited plutonium remained on the dishes. For each sample, the leachates were combined, evaporated to near dryness on a hot plate, taken up in 80 ml 9 mol/l HCl and the plutonium was again separated as described in Reference 16 by anion exchange on Bio Rad AG 1  $\times$  4, 100–200 mesh. The sample

effluent from the column in 9 mol/l HCl, as well as the total of 50 ml of 9 mol/l HCl washings, were successively combined, evaporated to near dryness, fumed with 0.5 ml of conc.  $\text{H}_2\text{SO}_4$ , and electrolyzed as described for the plutonium samples<sup>16</sup> to obtain the americium samples, which were then counted by  $\alpha$ -spectrometry.

### *Blank values*

Blank samples were prepared by processing the same amounts of reagents and tracers as given by the procedures described above. Total blank count rates were on the average  $1.64 \pm 0.69$  counts per minute in the low level proportional counter (six individual determinations). The a priori lower limit of detection at 95% confidence level,<sup>20</sup> based on the standard deviation of the total blank, on an average overall plutonium yield of 73% (after the second plutonium separation) and on a counting time of 1000 minutes is 10 mBq of  $^{241}\text{Pu}$  in the proportional counter.

## **Results and discussion**

Table 2 shows the results obtained from aerosol and deposition samples collected fortnightly and monthly, respectively, at our laboratory site at Neuherberg near Munich at the time of the Chernobyl accident. As indicated already in the introduction, these samples can be expected to provide an especially sensitive test of the entire chemical separation and proportional counting procedure, compared to samples contaminated only by global fallout, because interfering activities are much higher. The activity of e.g.,  $^{137}\text{Cs}$  was 9–10 kBq in the April 1986 deposition samples, while the maximum monthly  $^{137}\text{Cs}$  activity observed before, i.e., since the start of our measurements in 1970, was 4.8 Bq. The activity of  $^{239+240}\text{Pu}$ , however, was 26–46 mBq per deposition sample in April, 1986, which is comparable to the plutonium fallout activity observed by us during the 1970's.<sup>13</sup> After the first separation by anion exchange, some counts per minute to 100 counts per minute of residual  $\beta$ -activity remained in the samples, when counted through a filter paper absorber in an end-window proportional counter. However, after the additional lanthanum fluoride precipitation and TTA extraction, less than 0.1 counts per minute remained. This is low compared to the  $^{241}\text{Pu}$  net count rates of 15–65 counts per minute. A simple repetition of the first anion exchange separation did not remove the interferences sufficiently.

The main part of the interfering activity after the first anion exchange separation is due to the ruthenium nuclides, as shown by a  $\gamma$ -spectrometric measurement with a germanium detector. A fraction of less than 0.1 per cent of the total ruthenium had passed the initial plutonium separation; moreover, in a subsequent measurement with a

Table 2  
 $^{241}\text{Pu}$  activities (Bq) in air and deposition samples taken  
 at Neuherberg after the Chernobyl accident, determined with the  
 low level  $\beta$ -counter and via  $^{241}\text{Am}$  build-up

Sample from-to	$^{241}\text{Pu}$ (Bq)	
	Via $\beta$ -counting	Via $^{241}\text{Am}$
<i>Air</i>		
01.4. - 15.4.86	$\leq 5.4 \cdot 10^{-3}$	$\leq 0.19$
15.4. - 30.4.86	$2.09 \pm 0.02$	$1.88 \pm 0.20$
30.4. - 15.5.86	$8.26 \pm 0.08$	$8.08 \pm 0.67$
15.5. - 01.06.86	$\leq 5.6 \cdot 10^{-3}$	$\leq 0.20$
01.6. - 15.6.86	$\leq 6.2 \cdot 10^{-3}$	$\leq 0.21$
<i>Deposition</i>		
28.2. - 01.4.86	$\leq 5.4 \cdot 10^{-3}$	$\leq 0.20$
01.4. - 01.5.86	$3.64 \pm 0.02$	$3.60 \pm 0.27$
01.5. - 02.6.86	$1.02 \pm 0.01$	$0.87 \pm 0.10$
02.6. - 30.6.86	$\leq 5.9 \cdot 10^{-3}$	$\leq 0.23$

Reference date: May 1, 1986.

Errors: One standard deviation from counting statistics.

Table 3  
 Specific  $^{241}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  activities in air ( $\mu\text{Bq} \cdot \text{m}^{-3}$ )  
 and deposition ( $\text{mBq} \cdot \text{m}^{-2}$ ) at Neuherberg after the Chernobyl accident

Sample from-to	$^{241}\text{Pu}$	$^{239+240}\text{Pu}$	$^{137}\text{Cs}$
<i>Air</i>			
01.4. - 15.4.86	$< 0.16$	$< 0.026$	1.12
15.4. - 30.4.86	$39.3 \pm 0.4$	$0.61 \pm 0.04$	$156 \cdot 10^3$
30.4. - 15.5.86	$178 \pm 1$	$2.43 \pm 0.18$	$776 \cdot 10^3$
15.5. - 01.6.86	$< 0.11$	$< 0.023$	1240
01.6. - 15.6.86	$< 0.28$	$< 0.046$	290
<i>Deposition</i>			
28.2. - 01.4.86	$< 9.0$	$< 0.74$	0.14
01.4. - 01.5.86	$6065 \pm 28$	$79.5 \pm 2.3$	$17.4 \cdot 10^6$
01.5. - 02.6.86	$1697 \pm 11$	$25.2 \pm 1.6$	$2.3 \cdot 10^6$
02.6. - 30.6.86	$< 9.8$	$1.3 \pm 0.4$	190

Reference date: May 1, 1986.

Errors: One standard deviation from counting statistics.

low energy photon detector, some Te X-rays were observed and ascribed to  $^{125}\text{Sb}$ . The presence of traces of Ru and Sb is not unexpected from the anion exchange behavior of these elements.

The variation of the specific  $^{241}\text{Pu}$  activity with time shown in Table 3 is parallel to the variation of the many other radionuclides observed after the Chernobyl accident at Neuherberg,<sup>21,22</sup> thus indicating their common origin from the damaged reactor. Therefore, we have included in Table 3 also the temporal variation of  $^{137}\text{Cs}$ , which reflects the general variation of the bulk of longer-lived Chernobyl debris.

### *Quality control*

Unfortunately, there seems to exist no environmental standard reference material with a certified  $^{241}\text{Pu}$  content. For environmental SRM's available from NIST and IAEA only the  $\alpha$ -emitting plutonium isotopes are listed. Therefore, to compare the results obtained by proportional counting with those of an independent method, we used the build-up of the daughter nuclide  $^{241}\text{Am}$ , which has been previously reported.<sup>1,2</sup> The results are included in Table 2. The comparison shows satisfactory agreement between the two methods for all samples with  $^{241}\text{Pu}$  activities above the detection limit. A highly significant correlation was found between the  $^{241}\text{Pu}$  activity values determined by proportional counting and by  $^{241}\text{Am}$  build-up ( $r = 0.9997$ ;  $p < 0.1\%$ ). An application of the t-test showed no significant difference at the 95% level between the slope of the regression line (0.9986) and the ideal value of 1, and between the offset on the y-axis (-0.14) and the ideal value of 0.

### *Activity ratios in Chernobyl fallout*

From Tables 2 and 3,  $^{241}\text{Pu}/^{239+240}\text{Pu}$  activity ratios in Chernobyl debris can be calculated. Averaged over all samples with  $^{241}\text{Pu}$  activities above the detection limit, a mean of  $70 \pm 6$  is obtained with the proportional counting procedure, and of  $66 \pm 9$  via  $^{241}\text{Am}$  build-up, with no difference detectable between these means by the t-test at the 95% confidence level. There are few literature data available on the  $^{241}\text{Pu}/^{239+240}\text{Pu}$  ratio in fallout from Chernobyl. An overall value of 83 can be calculated from the Soviet data specifying the activities released from the reactor.<sup>23</sup> The few data on this ratio in long range Chernobyl fallout, reported directly as ratios or inferred from concentration data, scatter from 26 to 102.<sup>24-28</sup> This points possibly to an inhomogeneity of Chernobyl debris with respect to this ratio.

From recent mass-spectroscopic data of KRIVOKHATSKY et al.<sup>29</sup> for soil samples and average samples of fuel grains collected at a distance from 0.5 to 12 km from Chernobyl,  $^{241}\text{Pu}/^{239+240}\text{Pu}$  ratios of 67, 60 and 53 are calculated. These values are

similar to those of the present work. It is remarkable that also the ratios  $^{238}\text{Pu}/^{239+240}\text{Pu}$ , namely 0.42, and  $^{242}\text{Cm}/^{239+240}\text{Pu}$ , namely 12, found at Neuherberg in Chernobyl fallout samples,<sup>22</sup> are very similar to the near Chernobyl values of KRIVOKHATSKY et al., namely 0.36–0.48 and 9.1–19.2, respectively.<sup>29</sup> From this it can be concluded that no major isotopic fractionation of the actinides occurred between these two places.

#### *Advantages and disadvantages of the method*

Compared to the well-known method based on the  $^{241}\text{Am}$  build-up, the most important features of the proportional counting are as follows.

While the  $^{241}\text{Am}$  determination is based on the daughter nuclide, the measurement by proportional counting is direct. This means, that the results are practically immediately available, while, especially at the low environmental plutonium levels, a build-up time of 1–2 years is often required to accumulate sufficient  $^{241}\text{Am}$ . Even after 4 years, the activity ratio  $^{241}\text{Am}:^{241}\text{Pu}$  is only 1:166.

$^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Pu}$  can be determined in subsequent measurements from the same counting source. In contrast, in analysis via  $^{241}\text{Am}$  build-up, it is advisable, although not absolutely necessary, to separate the ingrown  $^{241}\text{Am}$  by radiochemical methods to avoid interference by  $^{238}\text{Pu}$ , which is present in most samples. Thus, the statistical counting error for  $^{241}\text{Am}$ , which has approximately the same  $\alpha$ -energy as  $^{238}\text{Pu}$  (Table 1), is minimized.

The lower limit of detection of  $^{241}\text{Pu}$  in proportional counting is about 10 mBq, based on the conditions of the present study, while it is about 200 mBq via  $^{241}\text{Am}$  build-up, under these conditions, based on a lower limit of detection for  $^{241}\text{Am}$  of 1 mBq in  $\alpha$ -spectrometry.

The proportional counting requires a high degree of purity of the counting samples, due to the generally nonspecific character of  $\beta$ -radiation. In contrast, the determination via  $^{241}\text{Am}$  is very specific and there is an additional identification by radiochemical separation. In fact, the chemical purity of the samples, due to interfering radionuclides and stable elements, leading to an increased self absorption, is the most critical point in the proportional counting procedure described.

The equipment for  $\alpha$ -spectrometric determination of  $^{241}\text{Am}$  is available in many laboratories dealing with analytics of radionuclides in the environment. The proportional counting needs special equipment, but this can be achieved by modifications of commercially available parts.



## References

1. H. D. LIVINGSTON, D. L. SCHNEIDER, V. T. BOWEN, *Earth Planet. Sci. Lett.*, 25 (1975) 361.
2. E. HOLM, *Environ. Int.*, 14 (1988) 363.
3. M. HAKANEN, T. JAAKKOLA, H. KORPELA, *Nucl. Instr. Meth.*, A223 (1984) 382.
4. G. G. HANDS, B. O. B. CONWAY, *Analyst*, 102 (1977) 934.
5. K. G. DARRAL, G. C. M. HAMMOND, J. F. C. TYLER, *Analyst*, 98 (1973) 358.
6. G. T. COOK, R. J. ANDERSON, *Radioanal. Nucl. Chem. Letters*, 154 (1991) 219.
7. W. R. SCHELL, C. E. VICK, E. A. WURTZ, *Methods of Low-Level Counting and Spectrometry*, IAEA, Vienna 1981, p. 125.
8. P. W. KREY, E. P. HARDY, C. PACHUCKI, F. ROURKE, J. COLUZZA, W. K. BENSON, *Transuranium Nuclides in the Environment*; IAEA, Vienna, 1976, p. 671.
9. K. O. BUESSELER, I. E. HALVERSON, *J. Environ. Radioactivity*, 5 (1987) 425.
10. W. MCCARTHY, M. T. NICHOLLS, *J. Environ. Radioactivity*, 12 (1990) 1.
11. J. C. DALTON, B. J. McDONALD, V. BARNES, *Radioisotope Sample Measurement Techniques in Medicine and Biology*, IAEA, Vienna, 1965, p. 347.
12. H. J. GALE, L. H. J. PEAPLE, Report AERE – R-4113, Harwell, 1962.
13. H. HÖTZL, G. ROSNER, R. WINKLER, Report GSF – S-956, Neuherberg near München, 1983.
14. K. BUNZL, W. KRACKE, *Sci. Total Environ.*, 63 (1987) 111.
15. K. BUNZL, K. HENRICH, W. KRACKE, *Assessment of Radioactive Contamination in Man*, IAEA, Vienna, 1985, p. 541.
16. G. ROSNER, H. HÖTZL, R. WINKLER, *Fresen. Z. Anal. Chem.*, 338 (1990) 606.
17. R. WINKLER, G. ROSNER, *Nuclear Instr. Methods*, A 274 (1989) 359.
18. H. HÖTZL, R. WINKLER, *Nuclear Instr. Methods*, A 223 (1984) 290.
19. W. KRACKE, K. BUNZL, *Radiochem. Radioanal. Letters*, 42 (1980) 77.
20. J. HARLEY (Ed.) *HASL Procedures Manual HASL-300 Health and Safety Laboratory*, New York, 1972, D-08-01.
21. H. HÖTZL, G. ROSNER, R. WINKLER, *Radiochim. Acta*, 41 (1987) 181.
22. G. ROSNER, H. HÖTZL, R. WINKLER, *Environ. Int.*, 14 (1988) 331.
23. Summary Report on the Post-Accident Review Meeting on the Chernobyl Accident; *Safety Series Nr. 75-INSAG-1*, International Atomic Energy Agency, Vienna, 1986.
24. K. BUNZL, W. KRACKE, *J. Radioanal. Nucl. Chem.*, 138 (1990) 83.
25. E. HOLM, J. RIOSECO, H. J. PETERSON, *J. Radioanal. Nucl. Chem.*, 156 (1992) 183.
26. G. BARCI-FUNEL, J. DALMASSO, G. ARDISSON, *J. Radioanal. Nucl. Chem.*, 156 (1992) 83.
27. O. LILJENZIN, M. SKALBERG, G. PERSSON, T. INGEMANSSON, P. O. ARONSSON, *Radiochim. Acta*, 43 (1988) 1.
28. R. BRODA, R. KUBICA, Z. SZEGLOWSKI, K. ZUBER, *Radiochim. Acta*, 48 (1989) 89.
29. A. S. KRIVOKHATSKY, Yu. V. DUBASOV, E. A. SMIRNOVA, N. V. SKOVORODKIN, V. G. SAVONENKOV, B. M. ALEXANDROV, E. L. LEBEDEV, *J. Radioanal. Nucl. Chem.*, 147 (1991) 141.