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RADIOAKTYWNOŚĆ ALFA W GORĄCYCH CZĄSTKACH Z CZERNOBYLA

АЛЬФА РАДИОАКТИВНОСТЬ В ЧАСТИЦАХ ИЗ ЧЕРНОБЫЛЬСКОГО ОСАДКА

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The alpha radioactive component of hot particles from the Chernobyl fallout was analysed for cases studied previously by gamma spectroscopy. Correlations established from the absolute alpha activity determination and high resolution analysis provided information on actinides release during accident and on some aspects of the Chernobyl reactor fuel composition.
Unexpected features revealed during the analysis of one specific particle are presented.

Przeprowadzono analizę alfa radioaktywnosci dla gorących cząstek badanych uprzednio metodami spektroskopii gamma. Bezwzględne pomiary aktywnosci alfa i analiza składu izotopowego pozwoliły ustalić korelacje prowadzące do wniosków o emisji aktynidów w awarii czernobylskiej i niektórych aspektach składu paliwa reaktora. Przedstawiono wyniki szczegółowej analizy cząstki, dla której zaobserwowano specyficzne własnosci.

Проведен анализ альфа радиоактивной компоненты для горячих частиц с осадка после чернобыльской аварии, которые прежде были исследованы методами гамма спектроскопии. Измерения абсалютных альфа активностией и анализ изотопического состава дали сведения о эмисии актинидов во время аварии, а также о некоторых аспектах связанных с чернобыльским реактором. Детально представлен анализ одной частицы, проявились некоторые интересные явления.

1. Introduction

Early announcement [1] of the presence of hot particles in the Chernobyl radioactive fallout was followed by a number of detailed studies performed in several european laboratories. Reported results [2-7] yield rather consistent picture regarding the origin and bulk properties of analysed particles. In our raport [2] we have presented results and conclusions obtained from the gamma spectroscopy analysis of 65 species collected in Poland. We have then indicated a parallel work on alpha spectroscopic analysis of hot particles referring to later publication. The present work summarizes the results obtained in nearly two year studies in this respect. It has been early recognized and observed [5,2] that the particles containing non-volatile radioisotopes (Ce,Zr) are predominantly small fragments of the reactor fuel with abundant alpha radioactive products formed by the neutron activation process. In the present work the alpha radioactive content of particles was studied for cases analysed previously by gamma spectroscopy. Thus knowing from the measured cerium and ruthenium isotope ratios the history of a specific fuel element, from which the particle has originated it was possible to establish some general correlations allowing to gain further information on particle formation process and behaviour of actinides in extreme conditions of the accident. Another objective was to check earlier conclusions [2] on the reactor fuel composition, as well as more practical application concerning the direct estimate of the alpha radioactive component activity in the fallout affecting the environment.

2. Remarks on properties of hot particles.

It has been generally accepted that the name "hot particles" denotes small radioactive fragments, which with rather high deposition rate contributed to the radioactive fallout, and were due to large specific activity distinguishable and separable from the continuous radioactive back-ground. The results obtained

by various groups show consistently the presence of two main categories of particles:

- monoelemental predominantly containing ruthenium radioisotopes
- fuel fragments including the non-volatile fission products with mostly depleted volatile elements, and varying amount of ruthenium.

The frequency of occurrence reported by different authors [3,2,6] is obviously sensitive to the detection threshold during a search; the high threshold strongly favours by order of magnitude more active ruthenium particles. Nevertheless the rapid decrease of the number of hot particles per area unit when moving along the radioactive cloud trajectory was immediately apparent indicating high deposition rate. This feature is naturally related to particle's size and density. Reported sizes varied considerably [3] and often were not correlated with the activity indicating presence of other core materials e.g. carbon, zirconium or silica. Devell has shown [3] that for ruthenium particles sizes of few micrometers measured for the radioactive part correspond to the diameters calculated from the measured activity assuming carrier free ruthenium as the only component. Recent studies [7] confirmed the metallic form of ruthenium with possible traces of oxygen localized only at the particle's surface. In our analysis we used the optical microscopy to determine sizes - obvious difficulty is how to isolate an object from the rest of a sample. For most cases our separation procedure was successful ending with an entity, which was clearly all of Chernobyl origin. Our confidence is based on several events when a large size particle of several hundred micrometers, after separation and microscopic analysis, could be fragmented into two or more parts sharing the initially observed activity. Of five ruthenium particles three did not show a partitioning of the activity, in other two cases the Ru radioactivity was divided unevenly into two (three in one case) parts. The fragmentation of the fuel like particle led to much more even partition of the radioactive content except for the ruthenium component which again did show strong clustering. Table I lists properties of three fragmented fuel-like particles determined from gamma measurements. For particles HS9 and M21, except for ruthenium

content all properties of two fragments are the same as in the original particle, but for particle M2 two fragments seem to arise from fuel of different burnup indicated by different cerium and ruthenium isotope ratios. The latter case might represent example of particle formed in time of accident by embedment of different fuel rod fragments into some nonradioactive material.

Table I. Properties of fragmented fuel-like particles established from gamma measurements (see text).

Code name Paricle	Total activity [Bq]	$\frac{^{144}Ce}{^{141}Ce}$	$\frac{^{106}Ru}{^{103}Ru}$	$\frac{^{103}Ru}{^{141}Ce}$	$\frac{^{95}Zr}{^{141}Ce}$
M2	313	0.44(1)	0.07(1)	0.50(2)	0.82(2)
a	240	0.38(3)	0.07(1)	0.56(2)	0.82(2)
b	45	0.66(4)	0.22(6)	0.37(3)	0.89(6)
M59	1537	0.51(1)	0.09(1)	0.31(1)	0.79(1)
a	380	0.54(2)	0.08(1)	0.46(3)	0.81(3)
b	913	0.53(3)	0.09(1)	0.40(4)	0.83(6)
M21	394	0.76(1)	0.21(1)	0.45(1)	0.85(1)
a	240	0.75(4)	0.24(2)	0.69(3)	0.85(4)
b	130	0.71(4)	-	<0.04	0.85(5)

Sizes determined for particles studied in the scandinavian fallout [3] are much smaller and generally correspond to the size estimated from the observed activity. In our optical microscopy analysis we observed 19 particles of different shapes with sizes varying between 20 and 600 μm. The large size fuel like particles often did show a structure of thin regular plates, but some of them looked very similar to monoelemental ruthenium particles, which exclusively represented three-dimensional, porrous conglomerates of irregular crystals. In fact this surprisingly large size of objects travelling by air for many hundreds of kilometers led us to suggestion that the bulk part of nonradioactive material of ruthenium particles were fragments of the reactor graphite moderator. The question is related to still open problem of formation mechanism of monoelemental ruthenium

(molibdenum) particles. It is well known [8] that even quite large size inclusions of ruthenium and molybdenum are formed in an irradiated fuel, yet it is surprising that the bulk of ruthenium activity bound in particles has shown up in a highly purified form with no trace of other non-volatile elements activity. On the other hand most of the ruthenium activity was released from the reactor in a form of volatile oxides and we suggested the possible formation mechanism of some of pure ruthenium particles as deoxidation process of ruthenium oxides in contact with burning graphite moderator followed by immediate condensation of metallic ruthenium. We reinspected our data on $^{106}Ru/^{103}Ru$ isotope ratios, which show distinct values for particles originating from fuel rods of various burnup. For 37 cases of monoelemental ruthenium particles the values are distributed around the average of 0.235 with variance σ = 0.038. For 19 cases of fuel like particles with ruthenium content large enough to determine the isotope ratio the distribution is much more scattered yielding somewhat lower average of 0.208 but with significantly larger variance σ = 0.086.Similar tendency is observed for the data presented by the Warsaw group [6]. This could indicate partial mixing of ruthenium activity originating from different fuel rods prior to monoelemental particle formation, which would be compatible with suggested above formation mechanism. It has to be mentioned that the size (activity) of ruthenium particles was found to be not correlated with ruthenium isotope ratio, which also reflects the burnup time. In fact the largest observed particle of 308kBq activity has shown relatively low value of $^{106}Ru/^{103}Ru$ = 0.081(2). On the other hand the ruthenium clusters contained in fuel like particles undoubtfully represent inclusions formed in an operating fuel. Except for particle size there are two other properties distinguishing particles collected in Scandinavia from those analysed in Poland. First, as follows from systematics presented by Devell [3] the depletion of volatile cesium was significantly less prominent for scandinavian particles, second, the data on isotope ratios obtained by Robertson [4] show nearly perfect correlation of $^{106}Ru/^{103}Ru$ and $^{144}Ce/^{141}Ce$ activity ratios. This has not been a case for particles studied by us and we concluded

[2] that at least in some parts of the reactor core the fuel composition was significantly different from the claimed uranium fuel with 2% ^{235}U enrichment. In summary it is possible that the particles deposited in Poland originated from different part of the Chernobyl reactor than those transported to Scandinavia. The strong depletion of cesium would suggest that the local temperature of this part could be higher; a possible cesium surface adsorption effect, if any, could be only more significant for particles of larger size. In this context an interesting question was raised by Devell [3] whether very small size particles of non-volatile elements were present in the fallout, or otherwise whether any vaporization of these elements took place during the accident. Recently published results [9] of analysis of long lived activities present in air filters exposed in Kraków during first days after accident indicate that very small, but detectable amount of Ce and Zr (Nb) radioisotopes was continously present in the radioactive cloud. This activity can represent the vaporized Ce and Zr since it shows the same transport properties as the bulk activity of volatile radioisotopes.
The alpha spectroscopy analysis presented in the next sections yielded further puzzling observations.

3. Experimental procedure

The analysis of alpha emitters in hot particles involved two steps. First, the aim was to determine as accurately as possible the integral absolute alpha activity of a specific particle and correlate it with properties established in earlier gamma measurements [2]. In a second step thin sources were prepared to perform high resolution alpha spectroscopic studies in order to identify and determine relative yields of all radioactive actinides present in a particle. The measurements were performed in vacuum chamber using surface barrier silicon detectors of 25 keV resolution for 5.8 MeV α energy and active area diameters of 5 and 20 mm. Except for the absolute alpha activity measurements the sources were placed as close as possible to the detector since the activities were very low extending the measurement time

to ten days in some cases. The large size isolated hot particles were fragmented by pressing between two glass plates and the fragments were picked up from the glass with a scotch tape. Usually, the particles were easily breakable and a typical spectrum obtained from such a thick source is shown in Fig. 1. The counts corresponding to alpha particles are well separated from the low energy but much more intense electron pulses. Cases for which the spectrum indicated significant absorption were not considered in this part of analysis. Alpha yield was determined for the solid angle callibrated source to detector distance and the absolute ^{144}Ce activity was measured with the gamma detector checking again the $^{144}Ce/^{141}Ce$ isotope ratio. For most cases less than 20% of original particle activity was lost in the above described source preparation procedure. To obtain thin sources the scotch tape with fragmented particle was roasted on a platinum plate in a temperature of ~ 500°C. Next the material was washed out with hydrochloric acid and treated with aqua regia. Some particles were easily soluble, but in a number of cases only part of the radioactive material could be dissolved. Later the solution was evaporated and dry remains were dissolved in 0.1 N H_2SO_4. The solutions pH was adjusted with NH_4OH to value of ~ 3.6 and electroplating was performed using the polished platinum or stainless steel disk as a cathode according to procedure described by Yaffe [10].

4. Experimental results and discussion

4.1. The absolute alpha activity measurements.

For 20 fuel-like particles of different burnup history the absolute alpha activity could be determined. Five examples of monoelemental ruthenium particles prepared in a similar way for α measurement did not show any trace of alpha counts. On the other hand an extraordinary particle (code name M44 in [2]) of very short burnup time and showing exclusively cerium isotopes γ radiation did indicate easily measurable alpha activity. Thus it was natural to correlate the observed absolute alpha activity with Ce radioisotopes, specifically with the ^{144}Ce long lived

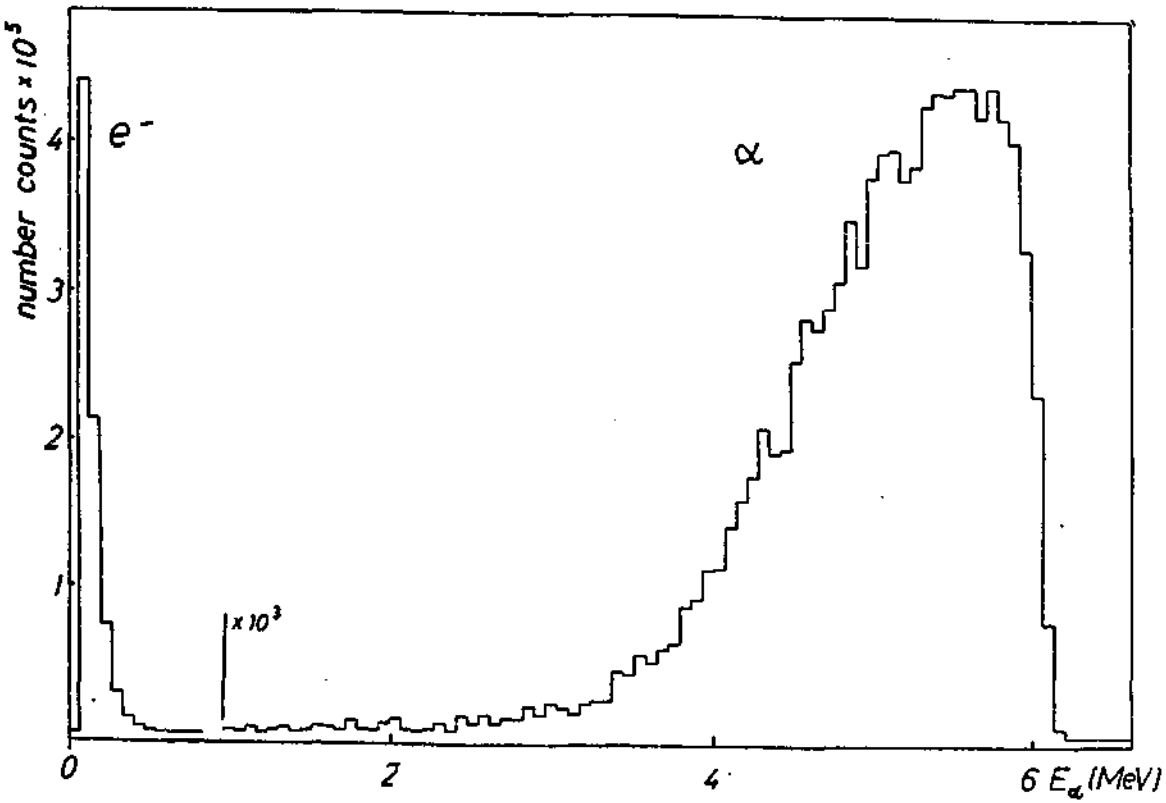

Fig.1. Example of a typical thick source alpha spectrum measured for the fragmented fuel-like particle.

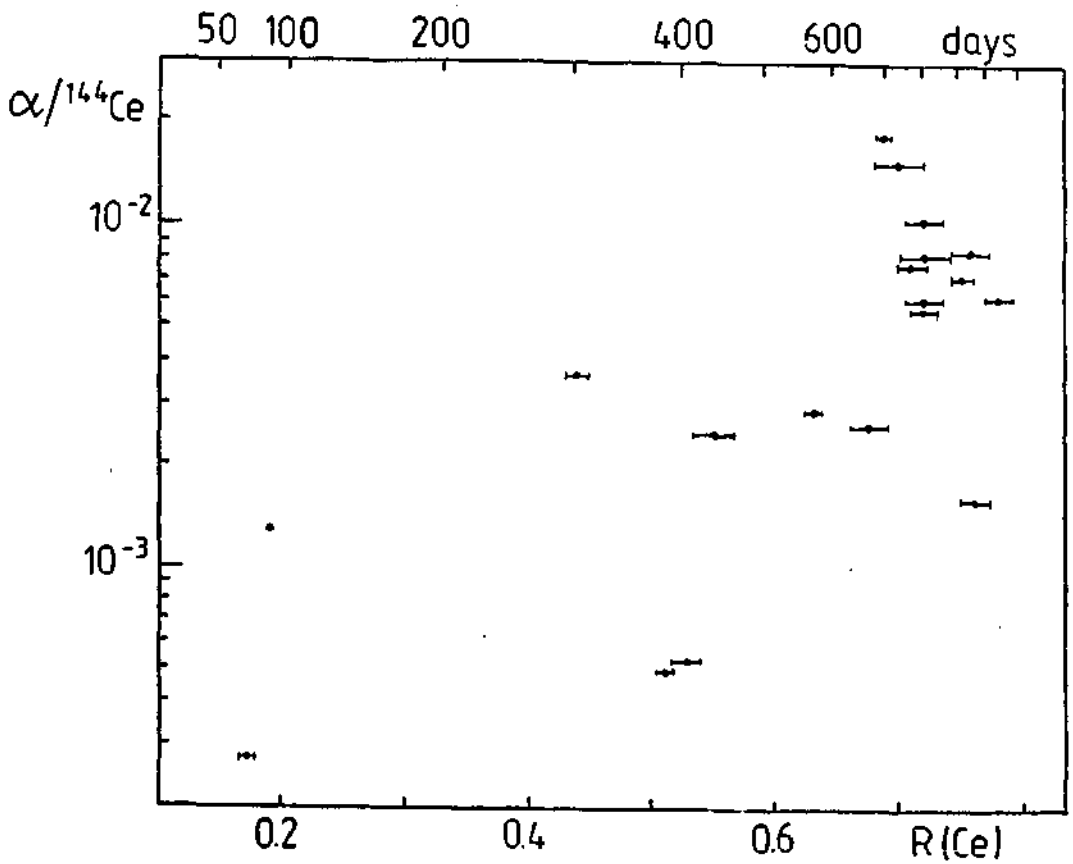

Fig.2. Correlation of alpha radioactive component of hot particles with cerium isotope ratio corresponding to fuel burn-up time.

($T_{1/2}$ = 284 d) isotope activity. Fig. 2 displays the obtained results and Table II gives numerical values for $\alpha_{tot}/^{144}Ce$ activity ratio including cerium isotope ratio established from the gamma analysis.

Table II. Integral alpha activity content determined for various fuel-like particles (as measured in October 1986).

Particle code name	$\frac{^{144}Ce}{^{141}Ce}$	α_{total}[Bq]$^{a)}$	$\frac{\alpha_{tot}}{^{144}Ce} \times 10^4$ $^{a)}$
HS2	0.175(4)	0.004(1)	2.8(7)
M44	0.190(2)	0.14	13
M2	0.441(10)	0.15(4)	37(10)
HS9	0.513(6)	0.094	5.0
M20	0.553(11)	0.089	5.4
M30	0.533(17)	0.53	25
HS14	0.632(5)	0.22	29
M24	0.675(17)	0.17	27
HS12	0.688(7)	1.37	187
HS11	0.698(19)	0.30	154
M3	0.709(12)	2.27	79
M29	0.716(10)	0.43	58
K01	0.720(15)	1.39	62
M1	0.722(28)	1.40	84
K02	0.722(15)	0.52	106
M12	0.750(8)	1.14	72
HS7	0.756(17)	0.47	85
M21	0.760(12)	0.14(4)	16(5)
M4	0.781(12)	1.52	63
M13	0.781(29)	0.59	63

$^{a)}$unless marked the uncertainty does not exceed 10%

All measurements were performed within first two weeks of October 1986, and the alpha activity was not corrected for possible decay. From the high resolution alpha analysis it turned out that at this date on average about 80% of the measured α counts

corresoond to the activity of ^{242}Cm ($T_{1/2}$ = 162.8 d), but for particles of short burnup time the long lived α activity prevails. Taking the average observed alpha rate one gets the relation allowing to estimate the total alpha activity from the known content of the ^{144}Ce radioisotope in the fallout.

A (α) = 1.1 × 10^{-2} A (^{144}Ce) at the end of April 1986, which is about 20% higher than the yield estimated from the core inventory [11]. After the decay of the ^{242}Cm component the remaining long lived activity will amount approx. to 15% of this initial value. In Fig. 2 the $\alpha/^{144}Ce$ activity ratio is displayed versus the cerium isotope ratio, which approximately determines the burnup time of a specific fuel element. Although the correlation in general is evident, the values are scattered showing features resembling the previously discussed [2] correlation of cerium and rutenium isotope ratios. Here the results may be affected by somewhat different behaviour of actinides and cerium in physico-chemical processes taking place during the accident, but the effect should not be drastic in view of earlier established [2] nearly perfect constancy of the relative yield of zirconium and cerium in various particles. The most natural explanation of scattered values of the $\alpha/^{144}Ce$ activity ratio leads again to conclusion on different fuel composition in various fuel rods of the reactor. Particles of comparable burnup time show the $\alpha/^{144}Ce$ ratio sometimes different by factor 5; it is interesting that the largest values were observed for particles with code names HS11 and HS12, for which especially large $^{106}Ru/^{103}Ru$ ratio has been observed [2] indicating predominant ^{239}Pu like fission.

4.2. High resolution alpha studies.

For number of particles the thin source preparation procedure worked very well and the obtained alpha spectra gave the identification and relative yields of various actinides present in an irradiated fuel. Fig. 3 shows example of α spectrum obtained for a particle of relatively long burnup time of approx. 800 days.

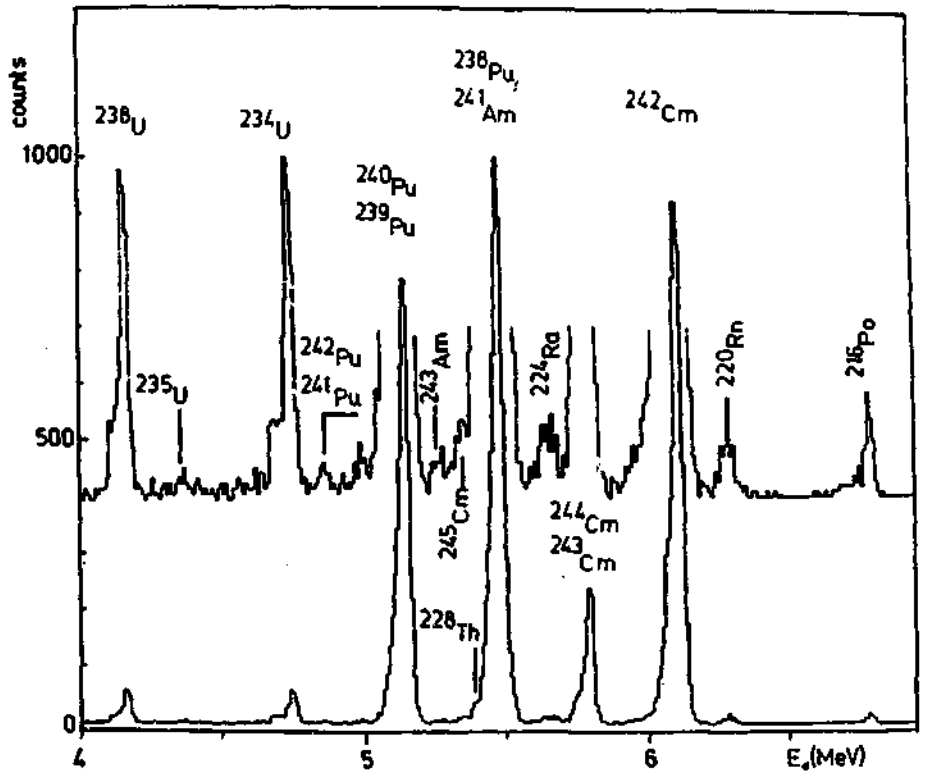

Fig.3. High resolution alpha spectrum measured for the particle of relatively long burn up time (see text). Spectrum is a sum of measurements performed in first three months of 1988. Upper part has shifted and expanded scale to emphasize weak lines. Results of quantitative analysis are given in Table III.

The activity values obtained from the quantitative analysis are given in Table III and compared with quantities from the estimated core inventory [11].

Table III. Composition of particle K01 established from gamma and alpha spectroscopy analysis (corrected for decay to 26 April (1986)).

Radioisotope	activity [Bq]	$\frac{A(\alpha,\gamma)}{A(^{144}Ce)}$	$\frac{A(\alpha,\gamma)}{A(^{144}Ce)}$ (core inv.)	mass (µg)
^{141}Ce	368(7)	1.39	1.75	3.5×10^{-7}
^{144}Ce	265(2)	1.00	1.00	2.2×10^{-5}
^{95}Zr	306(37)	1.15	-	2.1×10^{-7}
^{95}Nb	276(4)	1.04	1.53	3.5×10^{-7}
^{103}Ru	292(5)	1.10	1.56	2.4×10^{-7}
^{106}Ru	76(3)	0.29	0.62	6.1×10^{-7}
^{137}Cs	8.8(3)	0.033	0.091	2.7×10^{-6}
^{134}Cs	4.3(4)	0.016	0.059	0.9×10^{-7}
^{238}U	$9.6(2)\times10^{-7}$	3.2×10^{-5}	7.5×10^{-7}	0.69
^{234}U	$9.6(2)\times10^{-3}$	3.7×10^{-5}	7.8×10^{-7}	4.1×10^{-5}
$^{239,240}Pu$	$1.31(2)\times10^{-1}$	4.9×10^{-4}	6.4×10^{-4}	3.7×10^{-5}
$^{241}Am, ^{238}Pu$	$1.67(3)\times10^{-1}$	6.4×10^{-4}	6.2×10^{-4}	2.8×10^{-7}
$^{243,244}Cm$	$3.5(1)\times10^{-2}$	1.3×10^{-4}	-	1.2×10^{-8}
^{242}Cm	2.56(5)	9.8×10^{-3}	7.8×10^{-3}	2.1×10^{-8}

The agreement is fairly good and this selected example represents a particle of a typical fuel composition with uranium as predominant mass component (note discussion below). The change of relative yield of various alpha lines was followed within two years of measurements and gave $T_{1/2} = 163 \pm 3$ days for the 6.1 MeV line decay in perfect agreement with the ^{242}Cm half-life. The unresolved ^{238}Pu and ^{241}Am lines did show a systematic slow increase of intensity, which is naturally attributed to the growing in activity of ^{241}Am thus indicating a presence of the

^{241}Pu with activity roughly corresponding to the value expected in the core inventory. From the shape and centroid energy of the line marked as ^{243,244}Cm we deduce the predominant contribution of the ^{244}Cm isotope. Besides the ^{232}Th chain decay alpha lines, several other weak lines are marked in the spectrum of Fig. 3, but the identifications have to be considered as tentative. Whereas the observed traces of ^{235}U, ^{241,242}Pu and ^{243}Am lines are naturally expected, the more exotic ^{245}Cm line could be merely a statistical fluctuation. Similar quantitative analysis performed for alpha spectra of other particles gave results summarized in Table IV. Here the data are ordered with increasing particle's ^{144}Ce/^{141}Ce ratio listed in a second column. The values of the ^{239,240}Pu/^{144}Ce activity ratio given in a third column were obtained from the data of Table III with the ^{239,240}Pu content established from the high resolution spectra accounting properly for the ^{242}Cm component decay. These values are scattered around the average of 5.9×10^{-4} which is rather close to the calculated core inventory value. The scattering in general reflects observations made in the above discussion of the absolute alpha activity data, but more specific information is obtained when considering also other components present in alpha spectra, for which the relative yields are given in next columns of Table IV. The summed activity of ^{239}Pu and ^{240}Pu was used for normalization of other components since the unresolved lines of both isotopes appear in all measured spectra. The ^{238}Pu and ^{241}Am, as well as ^{243}Cm and ^{244}Cm are also unresolved doublets and the ^{242}Cm activity is corrected for the $T_{1/2}$ = 162.8 d decay to the 26 April 1986 accident time.

Comparing in Table IV the data obtained for particles of similar cerium isotope ratio one notices clear indication of varying fuel composition. The particles of comparable burnup time show different relative yields of various actinides. Again remarkable is the composition of particle HS12, which indicating unusually high yield of ^{239,240}Pu shows consequently significantly higher relative yield of the ^{242}Cm. Exceptionally the data obtained for particle HS7 suggest approx. factor 2 reduction of the ^{239,240}Pu component causing abnormally high relative yields of other actinides. The activity of unresolved ^{243,244}Cm is not given in

Table IV. Numerical results of high resolution analysis of alpha activities in hot particles.

Part, code name	^{144}Ce[a] / ^{141}Ce	$^{239,240}Pu$ / ^{144}Ce ($\times 10^{-4}$)	^{238}Pu, ^{241}Am / $^{239,240}Pu$	^{242}Cm[b] / $^{239,240}Pu$	$^{243,244}Cm$ / $^{239,240}Pu$	^{238}U[c] / $^{239,240}Pu$ ($\times 10^{-2}$)	^{230}Th / $^{239,240}Pu$
core inv.[d]	0.571	6.4	~1	12.2	-	0.12	-
HS2	0.175(4)	2.8(7)	-	-	-	-	-
M44	0.190(2)	12(1)	0.08(2)	0.08(3)	-	-	-
M2	0.38(3)	5(1)	0.2(1)	1.0(3)	-	-	-
	0.66(4)	5.3(7)	1.2(4)	9.5(8)	-	-	-
HS9	0.513(6)	2.5(1)	0.21(1)	2.1(1)	-	-	-
M20	0.533(11)	4.1(2)	0.19(1)	0.22(3)	0.030(5)	-	-
MC0	0.553(17)	5.5(3)	0.45(3)	6.9(3)	<0.04	-	-
HS14	0.632(5)	4.1(2)	0.67(7)	11(1)	0.09(3)	<5	0.46(8)
HS12	0.688(7)	14(1)	0.92(5)	29(1)	0.21(2)	-	-
M3	0.709(12)	8.2(3)	0.72(5)	16(1)	-	-	-
KD1	0.720(15)	6.5(2)	1.28(2)	20.4(2)	0.274(6)	7.0(3)	<0.005
M1	0.722(28)	5.6(7)	0.9(2)	25(5)	0.13(8)	-	0.13(7)
M22	0.735(7)	-	1.07(2)	12.5(2)	0.86(1)	4.1(4)	0.25(1)
M12	0.750(8)	5.3(3)	1.06(3)	23(1)	0.16(1)	<3	<0.03
HS7	0.756(17)	3.0(1)	1.4(1)	41(3)	0.46(4)	<2	0.21(4)
M13	0.781(29)	4.8(4)	0.72(9)	22(3)	0.25(5)	<4	0.30(6)
M4	0.781(12)	5.4(2)	0.80(3)	20(2)	0.11(2)	<2	<0.02

a) as determined from gamma measurement in ref.2.

b) corrected for $T_{1/2}$ = 162.8 decay to 26 April 1986.

c) taken as average from the ^{238}U, ^{234}U activity.

d) on April26. 1986. The ^{238}U activity calculated from information contained in Ref.[11].

the calculated core inventory [11], and the data of Table IV give quite scattered values for long burnup time particles. To some extent the production of these neutron rich Cm isotopes is sensitive to the distribution of the neutron flux within the reactor core, but the unusually high yield of $^{243,244}Cm$ observed for the particle M22 is rather striking. This particle has been a subject of particularly detailed analysis since it indicated other unexpected features as will be presented in a subsequent section.

For two particles the activity of ^{238}U could be detected and in both cases similar yield of the ^{234}U was observed; for five other cases upper limits are indicated. The amount of the ^{238}U observed in particles K01 and M22 exceeds by nearly two orders of magnitude the value expected for an irradiated fuel, and the absence of the ^{236}U activity suggests its outside reactor origin or particularly small concentration of the ^{235}U in this specific fuel elements. Also the presence of the ^{230}Th listed in the last column is surprising since being a product of the uranium natural decay it is not produced in any other process within the reactor based on uranium fuel. The traces of the ^{232}Th were also observed and one cannot exclude apriori the presence of the U, Th background, but its source would be completely ununderstood. Both Th and U activity appear irregularly and in uncorrelated way; the control runs in which the whole procedure was performed without any hot particle material, as well as separately for two ruthenium particles did not show traces of such activity.

4.3. Analysis of particle M22.

The gamma analysis did not reveal any particular features for the particle with code name M22 [2]. It indicated rather long burnup time with the cerium and ruthenium isotope ratio of 0.735(7) and 0.222(16) correspondingly, as well as significant depletion of ruthenium and cesium. In spite of very small size (~30μm) the absolute alpha activity could not be determined since the obtained alpha spectrum indicated strong absorption. On the other hand the particle was relatively easily soluble as noticed by observing the radioactivity during the standard procedure

applied in a preparation of thin source. The result of 1.5 hour electroplating was surprising yielding predominantly the activity of Pu isotopes with only traces of curium lines. The electroplating was repeated using new stainless disk cathode and readjusting the solution's pH to 3.6. The obtained new source revealed an increased relative yield of curium lines; identical procedure was repeated eight times until all radioactivity was extracted from the solution. Fig. 4 shows examples of alpha spectra measured for sources obtained from eight subsequent electroplatings. The observed systematic variation of different alpha emitters yield is quite puzzling especially when an identical procedure applied for numerous other particles did not show any Z dependence of electroplatings efficiency. It is hard to speculate on possible explanation of the observed effect since the difficulties in controlling electro-chemical process are particularly obvious for trace activities. Instead we tried to exploit the observed feature in an elemental analysis of particle's M22 composition. The gamma and alpha spectra for all eight sources were measured at callibratec positions with respect to detectors. The obtained variations of different radioisotope yield are displayed in Fig. 5. Of three observed gamma emitters the ^{144}Ce and ^{95}Zr (^{95}Nb) show similar behaviour with distinct peaks of efficiency in electroplatings No 3 and 6, but the yield of the ^{137}Cs is much more constant. On the other hand the actinides behaved quite differently, and among them the plutonium shows distinct property with nearly 95% of its total yield being extracted in first three electroplatings. The behaviour of plutonium supports conclusion that all particle material has been dissolved and merely electrochemical processes are responsible for observed feature of extrapolatings efficiency. Except for uranium all other actinides yield increases smoothly reaching maximum at electroplating No 6, but closer look indicated striking difference in behaviour of alpha lines attributed to ^{242}Cm and ^{243,244}Cm. This effect is reemphasized in the bottom right part of Fig. 5 showing the variation of relative yields of curium isotopes in subsequent electroplatings as measured in October 1987. Whereas at first two points the ratio ^{243,244}Cm/^{242}Cm shows values similar as observed in many other

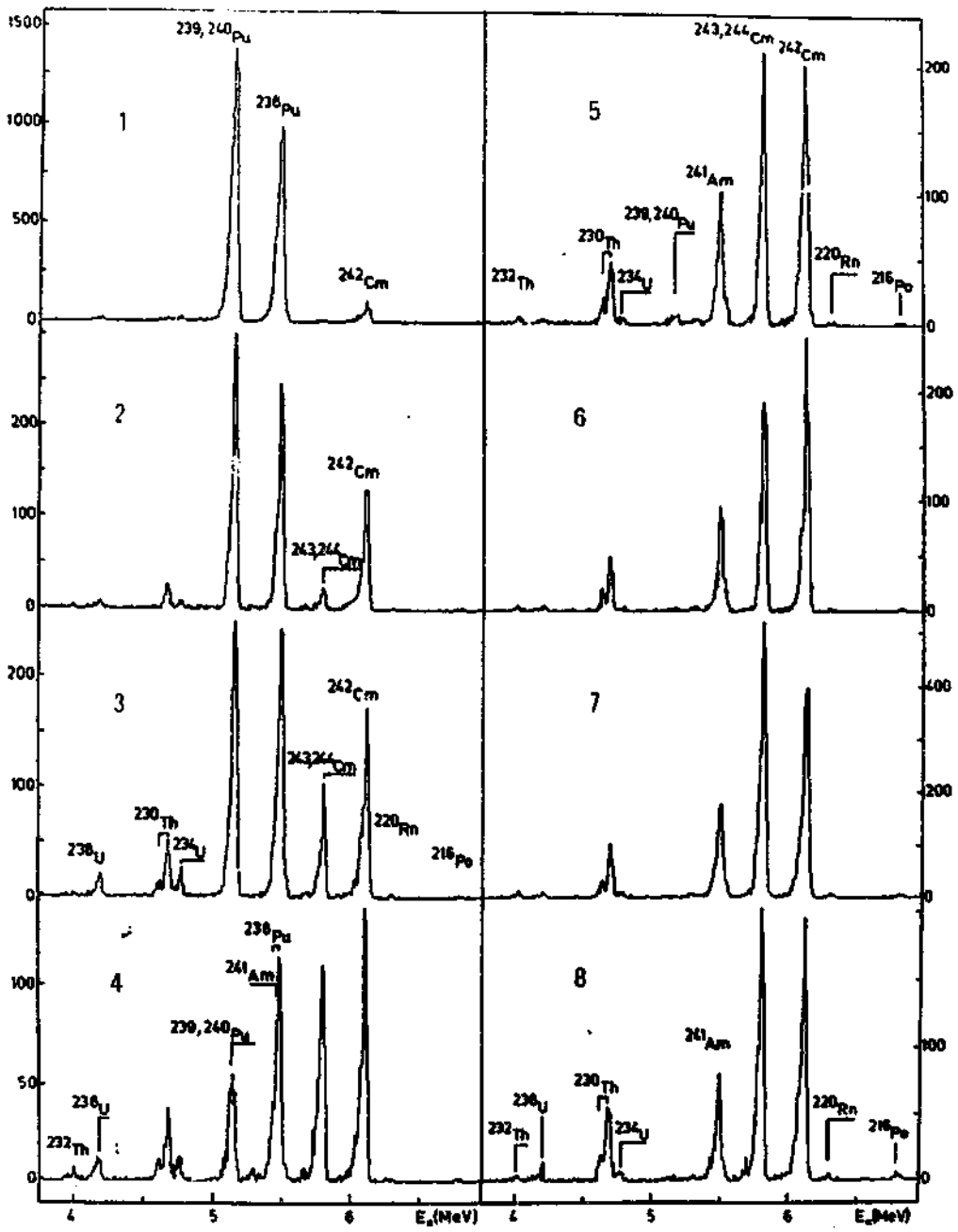

Fig.4. Examples of alpha spectra obtained from subsequent electro plantings of radioactive material of particle M22.

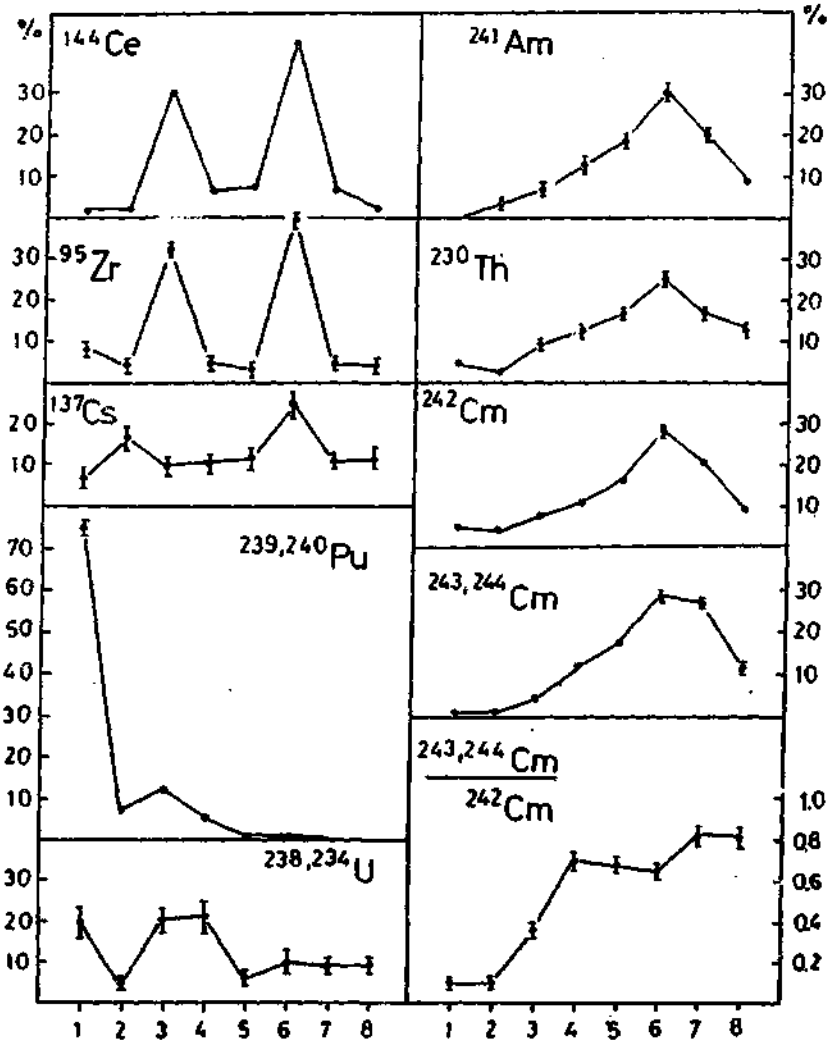

Fig.5. Yield of ectroplating process observed for various gamma and alpha emitters in analysis of particle M22.

particles, the yield of $^{243,244}Cm$ sharply increases starting with electroplating No 3. It is hard to conceive any possible mechanism leading to isotope effect, and natural suspicion was that some other alpha activity contributes to lines identified originally as $^{243,244}Cm$ or ^{242}Cm. To obtain additional information the variation in time of different aloha lines was followed for nearly two years of analysis of all eight sources. In first two sources containing predominantly plutonium activity a slow but systematic increase of the 5,5 MeV alpha line intensity was observed. Immediately after electroplating procedure this line is assumed to be pure ^{238}Pu activity and the increasing in time intensity is interpreted as growing in the ^{241}Am activity from the ^{241}Pu decay. The quantitative analysis allowed to deduce the value of the ^{241}Pu activity as given in Table V , which lists the established composition of actinides in particle M22 compared with values calculated from the core inventory [11].

Table V. Composition of actinides established in particle M22.

Radioisotope	Activity relative [a] to ^{239}Pu and ^{240}Pu		Mass relative to ^{239}Pu and ^{240}Pu
	Particle M22	Core inventory[11]	
$^{239,240}Pu$	1	1	1 [b]
^{238}Pu	0.67(3)	0.49	4.3×10^{-3}
^{241}Pu	102(15)	83	1.1×10^{-1}
^{241}Am	0.40(2)	-	1.3×10^{-2}
$^{243,244}Cm$	0.86(3)	-	1.4×10^{-2} [b]
^{242}Cm	12.5(3)	12.2	4.0×10^{-4}
^{236}U	0.046(2)	0.0012	1.5×10^{-4}
^{234}U	0.037(2)	-	6.5×10^{-4}
^{230}Th	0.25(1)	-	1.4
^{232}Th	0.023(3)	-	2.3×10^{-4}

[a] as determined for 26 April 1988.

[b] contribution of Pu isotopes taken from core inventory, for Cm isotopes assumed as 50% each.

The 5.8 MeV alpha line identified with the $^{243,244}Cm$ activity did not show any change in time of the intensity with respect to the long lived $^{239,240}Pu$. The decay of the 6.1 MeV ^{242}Cm line was followed by looking at its intensity relative to all other lines. The detailed measurements were performed only for selected alpha sources. The least square fit of these data gave the decay half lives of 146(9), 153(6), 162(4), 171(3) and 165(11) days for alpha sources from electroplatings No 1, 3, 5, 7 and 8 respectively. Compared to the 162.8 d ^{242}Cm half-life somewhat lower values at points 1 and 3 are well accounted for by the above discussed growth of the ^{241}Am activity, but the higher value observed for source No 7 could indicate a presence of very weak long lived component. However this result is inconclusive and clearly the bulk of the 6.1 MeV alpha line intensity has to be identified with the ^{242}Cm activity. In summary the two questions following from the analysis of the M22 particle remain unanswered. First - why the $^{243,244}Cm$ activity component is so large and second - why does it behave in different way in electrochemical processes than the ^{242}Cm. The only other actinide, which could be considered at the 5.8 MeV $^{243,244}Cm$ energy is the ^{249}Cf, but such consideration seems to be even more exotic.

The clarification of these questions exceeds the scientific scope of this paper and probably involvment of reactor experts could be helpful.

5. Conclusions.

The presented analysis of alpha active component in hot particles from the Chernobyl fallout gave the quantitative relation allowing to deduce the integral alpha activity from the known value of the ^{144}Ce gamma emitter activity present in the fallout. In general the actinides behaved in physico-chemical processes in a similar way as cerium, preserving the activity ratio, which is only by ~ 20% higher than estimated from the core inventory. For number of studied cases the various actinide yields correspond to the history of specific fuel elements deduced from earlier gamma analysis. For some particles the

results reemphasize the validity of earlier conclusion [2] on fuel composition. Detailed analysis of one specific particle posed unanswered questions, which may also relate to fuel composition. Comparison with results obtained in Scandinavia led to suggestion that hot particles deposited in Poland could originate from the different part of the Chernobyl reactor.

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