GAMMA SPECTROSCOPY ANALYSIS OF HOT PARTICLES FROM THE CHERNOBYL FALLOUT

By R. Broda

Institute of Nuclear Physics, Cracow*

(Received November 13, 1986)

Analysis of the gamma radiation was performed for 65 radioactive particles from the Chernobyl fallout. Isotopic ratios for Ce and Ru isotopes, as well as ratios of radioisotopes of various elements have been systematized and provided information concerning general features of processes leading to the hot particle formation.

PACS numbers: 28.90.+i

1. Introduction

Highly inhomogeneous distribution of radioactive fallout following the Chernobyl accident was one of the surprises which contributed to serious difficulties in early estimates of the situation at different locations in Poland.

On a macroscale a computer simulation based on meteorological information [1] reproduced rather well a general trend in radioactive cloud spread, but more detailed comparison with empirical data on ground level deposits show some significant differences with this result. For example, in Kraków area of southern Poland high ground level activity was observed reaching in some places up to 360 kBq/m^2, as determined on May 1-st [2]. A clear reason for this was a coincidence of intense rainfall and radioactive cloud passage. Such correlation was later established [3] and reported also at other places in Europe [4]. Thus precipitation is probably a simple explanation of large differences in a magnitude of radioactive fallout at various, sometimes not very distant locations.

Somewhat more puzzling are spots of radioactivity of much too small dimensions to be explained in this way. We observed in Masurian Lakes region in Poland spots as small as of 30 cm diameter, with nearly uniformly distributed activity of 20 times higher intensity of radiation than the background measured in a large area around it. The origin of these spots is not clear as yet, but they could arise as a result of successive erosion and wash-out by rains of hot particles similar to those investigated in the present work, but of larger size.

* Address: Instytut Fizyki Jądrowej, Radzikowskiego 152, 31-342 Kraków, Poland.

Probably the most interesting is a microscale inhomogeneity — the hot radioactive particles which have been quite early observed in the Chernobyl fallout. Such particles were first reported by Studsvik group [5] and later confirmed by physicists from Konstanz [4]. In the second half of May we have found few of such particles in the Masurian Lakes region, but detailed investigation started only one month later.

For the purpose of this work a hot particle is defined as an entity formed in the time of accident and found in the fallout as distinct concentration of radioactivity within single piece of very small size. A thorough investigation of these particles seems to be important for two reasons:

1. it may bring useful information on physical and/or chemical processes which took place during accident
2. establishing possible correlations between different radioactive elements may have practical application in speedy estimate of the amount of difficult for detection, but very important components in radioactive contamination.

As will be briefly discussed later we have found abundant alpha emitters contained in some hot particles [6], and there are reasons to expect sizable activity of strontium radionuclides — quantitative assignment for both of them is much more difficult than for radionuclides which emit gamma rays.

With these goals in mind an attempt was made to perform investigation of statistically significant number of hot particles. In all 65 hot particles were found, separated and subjected to examination using the gamma, X-ray and alpha-spectroscopy as well as optical microscopy and chemical analysis.

In the present work the result of the gamma spectroscopy investigation is presented. Results obtained with other techniques will be published separately.

2. Distribution and general properties of hot particles

Of 65 particles analysed in the present work 15 were found in the Kraków region and subjected to gamma ray measurements in July. The other 50 were collected in Mikołajki (Masurian Lakes region) in early September and measured within next two weeks. In most of the cases they were searched for on the ground using an ordinary Geiger-Mueller radiometer.

In north-east Poland hot particles were much more frequent than in the south, but distributed inhomogeneously. A systematic search in an open terrain in Mikołajki gave a very rough estimate of one particle per ten square meters (particle of more than 100 Bq activity at time of search). Similarly high density was observed in early May in Lublin region of east Poland, where they could be easily distinguished from rather low, in absence of rain, continuous background activity [7]. A search in Kraków region was more complicated because of intense background activity. An example of radioactive fallout composition is given in Table I (from Ref. [2]). It includes predominantly volatile reactor products, and observed mostly uniform distribution of Ru radionuclides suggested also for ruthenium its volatile oxide form. Whereas non-volatile reactor products such as cerium, niobium or zirconium were nearly absent, the appearance of gamma lines characteristic for these

TABLE I

Example of radioactive fallout composition in Kraków — Poland. A soil sample of 0.5 cm layer was measured 1-st of May (Ref. [2])

^{132}Te — 29.3 kBq/m^2	^{140}Ba — 2.5 kBq/m^2
^{132}J — 25.7 kBq/m^2	^{140}La — 2.4 kBq/m^2
^{131}J — 23.6 kBq/m^2	^{99}Mo — 1.7 kBq/m^2
^{129m}Te — 8.0 kBq/m^2	^{106}Ru — 1.3 kBq/m^2
^{103}Ru — 6.1 kBq/m^2	^{127}Sb — 0.8 kBq/m^2
^{137}Cs — 5.2 kBq/m^2	^{136}Cs — 0.7 kBq/m^2
^{134}Cs — 2.7 kBq/m^2	

TABLE II

Radioactive composition of gamma emitters in hot particles. Activity values are extrapolated to 26 April, 1986

	Particle 1	Particle 2	Particle 3	Particle 4
^{141}Ce	—	368(7)	558(2)	4(3)
^{144}Ce	—	265(2)	106.3(4)	1.4(8)
^{103}Ru	23790(150)	292(6)	1.6(4)	3306(7)
^{106}Ru	4860(45)	76.3(2.6)	—	743(2)
^{95}Zr	—	276(4)	1.3(3)	6(1)
^{95}Nb	—	306(33)	1.3(5)	544(4)
^{137}Cs	—	8.8(3)	0.16(3)	—
^{134}Cs	—	4.3(4)	—	—

radionuclides was always an indication of a hot particle presence in a measured sample. It could be then always traced down to a single small size entity, separable with some effort from the rest of a sample.

Gamma ray measurements indicated existence of two types of hot particles with distinctively different compositions, in accord with observation made by Studsvik groups [5]. Typical examples of radioactive composition are given in Table II in first two columns. It should be noted that in Table II and in the following, unless specified, all quantities related to hot particles, such as activity, isotopic ratios etc. are values extrapolated to the chain reaction stopping time taken as April 26-th 1.00 a.m. This is, all values are corrected for the decay which took place until the time of measurement.

An example of the first type particle gamma spectrum is shown in Fig. 1. Such particles contained highly pure radioactivity of ^{103}Ru and ^{106}Ru isotopes, with usually high activity ranging from 300 Bq up to 308 kBq measured in a single case, but typically several tens of kBq. As mentioned above, measurement were performed in July and September series. Consequently it was too late to detect any activity of ^{132}Te and ^{131}I isotopes, traces of which have been observed by Studsvik group. On the other hand the 27.7 day half-life of the ^{51}Cr radioisotope reported in [5] made this component easily accessible in the present measurements, but none of the ruthenium hot particles analysed here did show such activity. Of 65 analysed particles 37 belonged to this category.

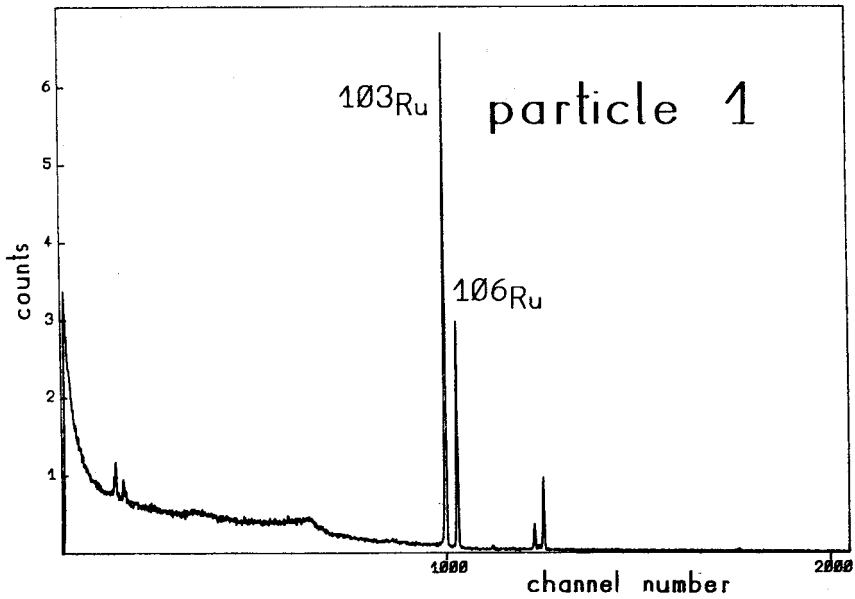

Fig. 1. Gamma spectrum of ruthenium particle measured September 19, 1986. Besides the lead X ray lines seen at low energies all other lines originate from the ^{103}Ru and ^{106}Ru decays

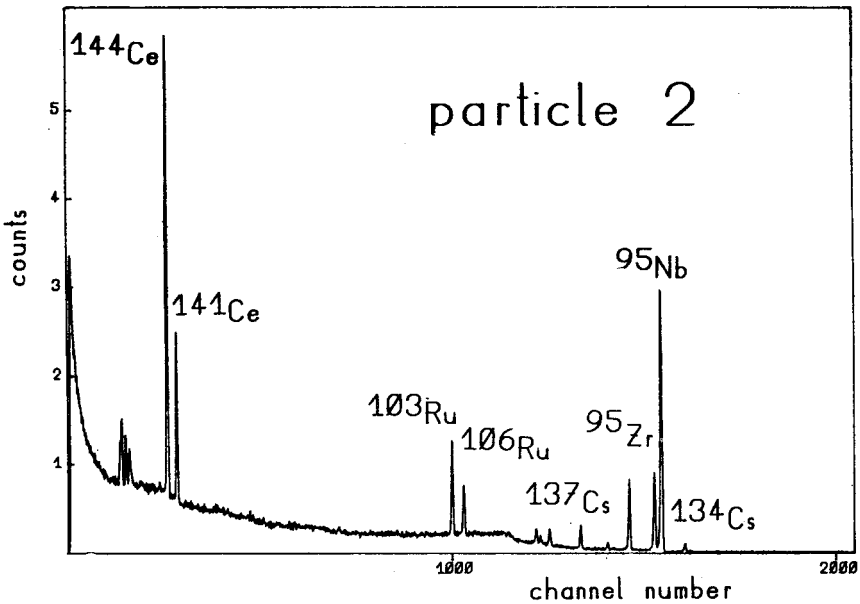

Fig. 2. Gamma spectrum of the second type particle M4 (see Table IV) measured September 10, 1986

The second type particles show more complex composition and an example of typical gamma spectrum is displayed in Fig. 2. These particles contain roughly in similar proportions radioisotopes of ^{141}Ce, ^{144}Ce, ^{95}Zr and ^{95}Nb as well as varying contribution of ^{103}Ru, ^{106}Ru, and small amount of ^{137}Cs, ^{134}Cs isotopes. In July series of measurements the ^{140}Ba-^{140}La activity was observed in some species. Extrapolation to chain reaction stopping time gave values comparable to Ce isotopes activity, but never as dominant as quoted by Studsvik group [5] in their example. In contrast to ruthenium particles and in accord with early observation of the short-lived ^{239}Np activity [5] these particles show a presence of alpha emitters, of which the dominant ^{242}Cm, and significantly weaker alpha lines of ^{241}Am and ^{239}Pu were clearly established [6]. Thus it appears most likely that at least the radioactive parts of the second type particles are small fragments of the reactor fuel from which the volatile products mostly escaped. The total activity for gamma emitters present in these particles was ranging from 12 Bq to nearly 2 kBq not depending on the place where they were found. As an instance a particle of 220 Bq was separated from the stem of a single berry of black current, the one with 1.5 kBq activity was attached to a pine tree needle and the 12 Bq particle (5.5 Bq in time of measurement) was detected on a plant leaf.

Among all hot particles analysed in the present work there were two extraordinary examples. Their gamma spectra are shown in Figs. 3 and 4, and compositions are listed in Table II. The first one contained nearly pure activity of cerium radioisotopes and the second is a ruthenium particle with large contamination of the ^{95}Nb activity but drastically reduced ^{95}Zr and ^{141,144}Ce activities.

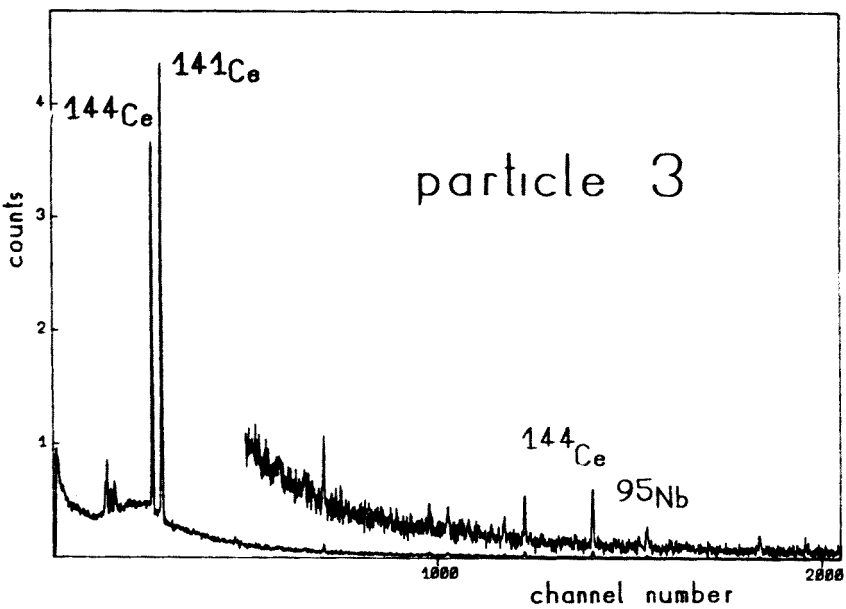

Fig. 3. Gamma spectrum of particle M44 (see Table IV) measured September 20, 1986. Unmarked lines relate to gamma background

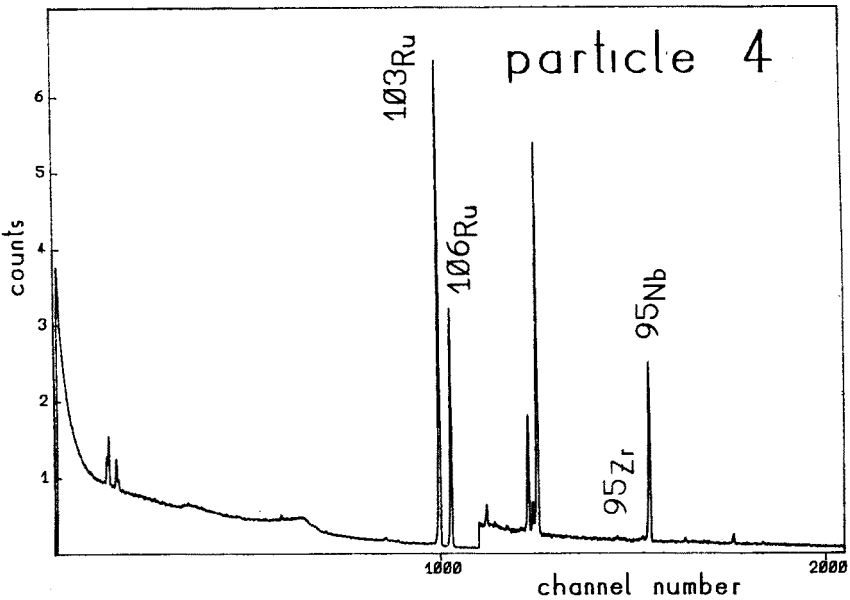

Fig. 4. Gamma spectrum of particle M39 (see Table IV) measured September 19, 1986. Most of lines seen in the spectrum belong to the ^{103}Ru, ^{106}Ru decays

The preparation of hot particles for optical microscopy is a tedious task and analysis is still in progress [6]. Few examples examined till now indicate that they are of surprisingly large size — up to several hundred micrometers, often visible with an unarmed eye. Having a size not correlated with activity they show a structure of easily breakable, mostly irregular crystals with indication of much smaller embedments, possibly similar to ball like structures observed at Studsvik [5]. They certainly consist of mostly nonradioactive material and probably some of them are the reactor graphite moderator pieces, direct evidence for it has not been obtained as yet.

3. Measurements and analysis

Gamma ray measurements were performed with the 40cc Ge(Li) detector of 2.5 keV resolution at 662 keV and calibrated for absolute efficiency. Analysed particles were always separated from the rest of a sample ensuring calibrated point-like geometry and absence of any significant absorption. Statistics collected in the gamma spectra was aimed to achieve the highest possible accuracy for activity determination of all radioactive products observed. As seen from gamma spectra of Figs 1–4 all lines used in analysis were well resolved allowing to use simplest procedure of peak area integration and linear background subtraction. Since this procedure was applied in an identical way for all measured samples, statistical errors determined accuracy to which samples may be compared with each other; uncertainty of activity absolute values is estimated as 5%. Properties of radionuclides used in analysis are listed in Table III.

TABLE III

Properties of radionuclides decay used in the analysis (Ref. [8])

Isotope	$T_{1/2}$	Strongest gamma line E[keV]	% per decay	Other gamma lines
^{141}Ce	32.5d	145.4	48.4	—
^{144}Ce	284.8d	133.5	11.1	80.1, 696[a], 814[a]
^{103}Ru	39.35d	497.1	86.4	557.1, 610.3
^{106}Ru	368d	511.9[a]	19.0	622.2[a], 873.7[a], 1050.5[a]
^{95}Zr	64d	756.7	54.6	—
		724.2	44.2	—
^{95}Nb	35d	765.8	99.8	—
^{140}Ba	12.79d	537.3	23.6	162.6, 328.8[a], 815.8[a]
		487.0[a]	43.0	
		1596.2[a]	95.5	
^{137}Cs	30.17y	661.7	85.0	—
^{134}Cs	2.06y	604.7	97.6	563.3, 569.3, 801.8
		795.8	85.4	

[a] transition from the daughter nucleus decay.

For ^{106}Ru activity determination the 512 keV gamma line was used rather than the much weaker 622 keV line, even if it was necessary to subtract a contribution from the unresolved 511 keV line present in the background spectrum. This correction was carefully determined and never exceeded 30%, for most of the cases being negligible. Quantities, which have been calculated from the measured and corrected for decay activities are: cerium and ruthenium isotope ratios, ratios for isotopes of various elements and total activity of all gamma emitters observed in a sample. Obtained values for 28 particles of the second type are listed in Table IV ordered with increasing ^{144}Ce/^{141}Ce ratio. For ruthenium particles the isotopic ratios were similar to those listed in Table IV but determined with higher accuracy since generally the activity was significantly larger.

4. Results and discussion

4.1. The ^{144}Ce/^{141}Ce and ^{106}Ru/^{103}Ru ratio

The isotope ratio for cerium and ruthenium radionuclides may be used for deduction of an average age of the reactor core. It has been estimated by other authors as 400 days from the ^{137}Cs/^{134}Cs ratio [5] and as (2.0±0.5) years from the ^{106}Ru/^{103}Ru ratio [4]. The average value of the ^{106}Ru/^{103}Ru and ^{144}Ce/^{141}Ce ratio determined for hot particles in the present work is 0.225 and 0.632 correspondingly, and both values are very much the same as values obtained in the analysis of continuous radioactive fallout, e.g. Ref. [2] and [4].

All four radionuclides are produced in a reactor fuel as a result of fission process of predominantly ^{235}U and ^{239}Pu nuclei. Whereas the ^{235}U fuel burns out, the amount of

TABLE IV

Numerical values obtained in the analysis of 28 particles of the second type. Values are corrected for decay and extrapolated to chain reaction stopping time (26 April, 86, 1.00 am)

Code name	$\frac{^{144}Ce}{^{141}Ce}$	$\frac{^{106}Ru}{^{103}Ru}$	$\frac{^{141}Ce}{^{95}Zr}$	$\frac{^{141}Ce}{^{103}Ru}$	$\frac{^{95}Nb}{^{95}Zr}$	$\frac{^{137}Cs}{^{141}Ce} \times 10^2$	$\frac{^{140}Ba}{^{141}Ce}$	Total activity Bq
HS2	0.175(4)	0.020(12)	1.75(4)	2.39(5)	0.46(5)	0.6(1)	1.02(7)	220
M44	0.190(2)	—	420(80)	350(90)	—	—	—	668
MO2	0.441(10)	0.070(12)	1.22(3)	2.01(6)	0.80(9)	1.0(1)	—	313
HS9	0.513(6)	0.092(15)	1.27(1)	3.24(6)	0.89(3)	1.3(1)	0.50(5)	1537
HS13	0.519(15)	—	1.19(3)	>60	0.88(7)	<0.1	0.4(1)	440
M20	0.533(11)	—	1.08(3)	>100	0.75(9)	<0.1	—	1137
M30	0.553(17)	0.017(80)	0.74(2)	7.6(1.3)	0.20(10)	0.9(2)	—	1008
HS8	0.556(15)	0.144(22)	1.24(2)	1.75(4)	0.96(4)	—	0.5(2)	109
HS3	0.600(33)	0.53(60)	1.23(5)	1.49(6)	1.07(12)	—	1.4(4)	41
HS14	0.632(5)	—	1.23(1)	>240	0.33(2)	0.23(5)	0.47(6)	388
HS4	0.65(9)	—	1.00(10)	1.19(11)	0.92(20)	—	—	12
M24	0.675(17)	0.232(2)	1.27(3)	0.078(2)	0.70(9)	<0.1	—	1792
HS5	0.690(31)	0.141(23)	1.25(5)	1.24(5)	1.15(10)	—	—	57
HS12	0.688(7)	0.341(10)	1.34(1)	1.16(1)	0.78(3)	<0.1	0.35(4)	461
HS11	0.698(19)	0.335(23)	1.36(4)	1.58(4)	0.81(7)	0.8(2)	<0.4	112
HS16	0.703(12)	0.127(18)	1.18(2)	2.09(5)	1.05(3)	2.2(2)	0.8(1)	138
M3	0.709(12)	0.242(11)	1.12(12)	1.81(5)	0.70(8)	2.4(1)	—	313
M29	0.716(10)	—	1.24(2)	>110	0.99(8)	0.29(5)	—	1370
KO1	0.720(15)	0.261(10)	1.33(4)	1.26(4)	1.11(12)	2.4(1)	—	1596
KO2	0.721(17)	0.281(11)	1.29(3)	1.33(4)	1.10(14)	4.5(2)	—	290
M1	0.722(28)	0.174(22)	1.16(5)	1.84(11)	0.96(41)	<0.2	—	939
HS6	0.722(34)	—	0.90(3)	>100	0.96(9)	—	—	47
M22	0.735(7)	0.222(16)	1.25(2)	5.35(16)	0.01(5)	0.54(3)	—	854
M12	0.750(8)	0.263(7)	1.18(2)	1.48(3)	0.94(6)	4.3(1)	—	927
HS7	0.756(17)	0.239(21)	1.30(3)	1.38(3)	0.92(10)	—	<0.2	368
M21	0.760(12)	0.213(13)	1.17(2)	2.22(6)	0.64(7)	3.0(1)	—	394
M4	0.781(12)	0.272(10)	1.14(2)	1.51(3)	1.05(8)	3.9(1)	—	1381
M13	0.781(29)	0.277(22)	1.19(5)	1.46(7)	1.08(19)	5.7(3)	—	532

^{239}Pu arising from the neutron capture on ^{238}U is increasing with time of the reactor operation. The mass number distribution for fission products is different for ^{235}U and ^{239}Pu fission, and consequently expected isotope ratios depend on proportion in which both fission events have happened. The ^{144}Ce/^{141}Ce yield ratio is by factor of 1.34 larger in the ^{235}U fission than in the ^{239}Pu case. The ^{106}Ru/^{103}Ru yield ratio is even more sensitive, and changes the opposite way with corresponding factor of 0.215. Rather complicated computer codes have to be used to predict variation of both isotopic ratios as a function of reactor core age. Besides taking into account also other possible fission processes, such as ^{238}U fission induced by fast neutrons, a complete knowledge of fuel composition and reactor flux characteristics is necessary. The analysis presented in Fig. 5, which uses simultaneously both cerium and ruthenium ratios is to large extent independent of this complication. Additionally the similarity of half-lives for short-lived and long-lived Ce and Ru isotopes mostly removes ambiguity related to detailed history of the reactors operation. Underlying assumption is that only the ^{235}U and ^{239}Pu fission takes place, and the values for fission yields are taken from 1977 Rider, Meek compilation [9].

The observed values of isotopic ratios fix equations between the core age and a parameter which describes the sharing of the fission events between ^{235}U and ^{239}Pu nuclei. Corresponding curves in Fig. 5 represent these equations and the crossing point matches both observed isotopic ratios. It determines with great accuracy 570 days as an average age of reactor core in which 43.3% of ^{239}Pu and 56.7% of ^{235}U fission events took place. It has to be emphasized that this result is valid for the reactor core as a whole and, as will

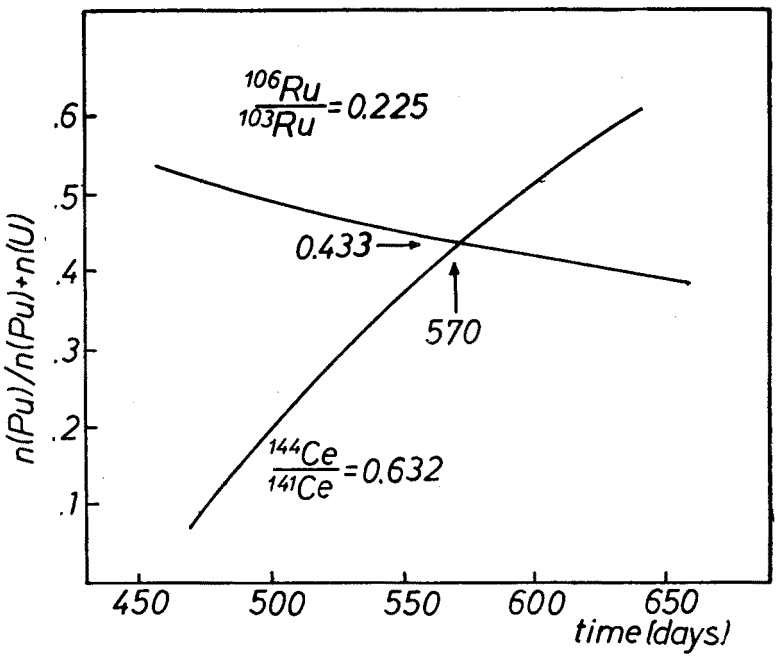

Fig. 5. Extraction of the average age of the reactor core (see text)

be seen in the following, the average isotopic ratios were obtained from many precisely determined but very different values.

Of all analysed particles for 28 the $^{144}Ce/^{141}Ce$ and for 56 the $^{106}Ru/^{103}Ru$ ratio could be determined with reasonable accuracy. Fig. 6 shows a histogram constructed from the obtained values of the $^{144}Ce/^{141}Ce$ ratio and it represents a distribution of this quantity in different fuel elements of the reactor core.

In somewhat more realistic way the same data are displayed in Fig. 7. Here, the summation was performed of gaussian functions representing each value with area normalized to unity and width equal to uncertainty. The distribution is then a differential one; such procedure was applied also in other cases discussed below.

From the emerging picture it is clear that the hot particles originated from different elements of the reactor fuel, with various history and no significant mixing has occurred during accident. In construction of the time scale in Figs 6 and 7 the earlier extracted average value of fuel composition was used. A small sensitivity of cerium isotopic ratio to varying in time fuel composition makes this approximation reasonable. The distribution is concentrated mostly around 800 days peak, but clearly some parts of the fuel have been operating for shorter time with two examples of time as short as 2.5 months. Fig. 8 displays similar distribution for ruthenium isotope ratio. Compared to distribution of Fig. 7 it is somewhat broader suggesting additional effect of fuel composition.

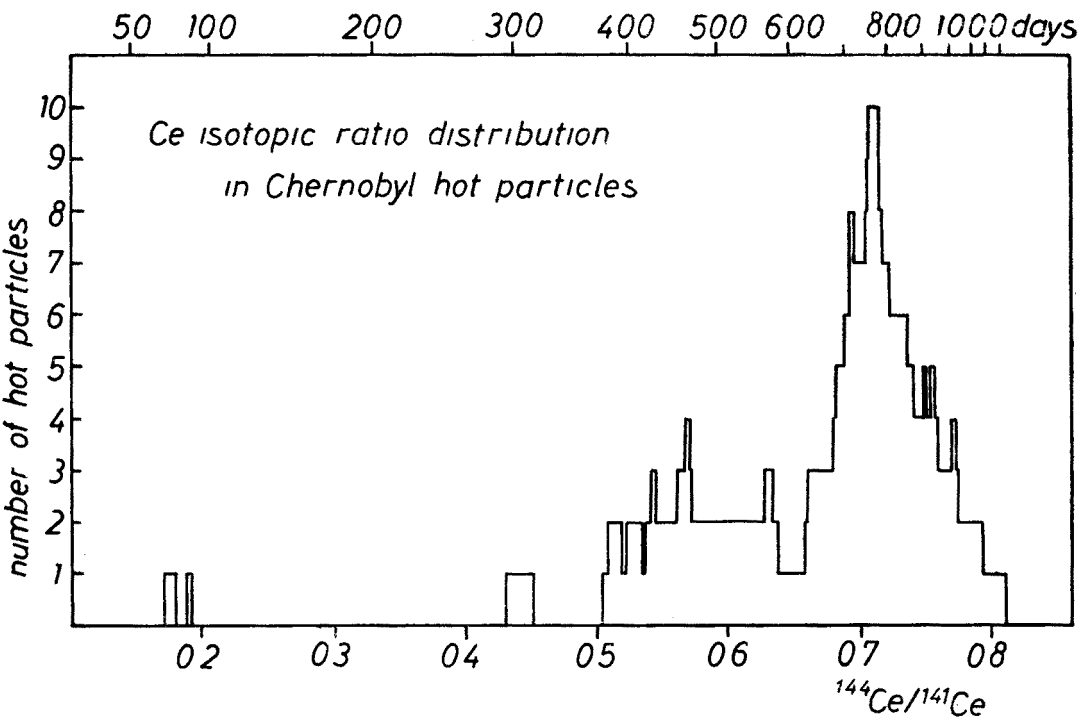

Fig. 6. Histogram of the cerium isotope ratio observed in hot particles

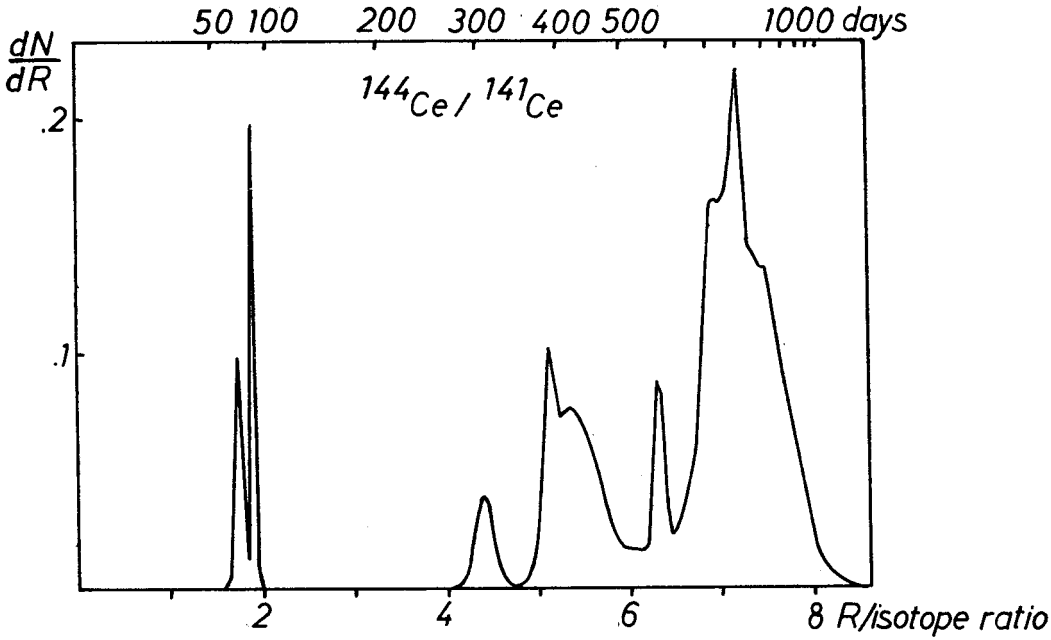

Fig. 7. Distribution of the $^{144}Ce/^{141}Ce$ isotope ratio in analysed particles

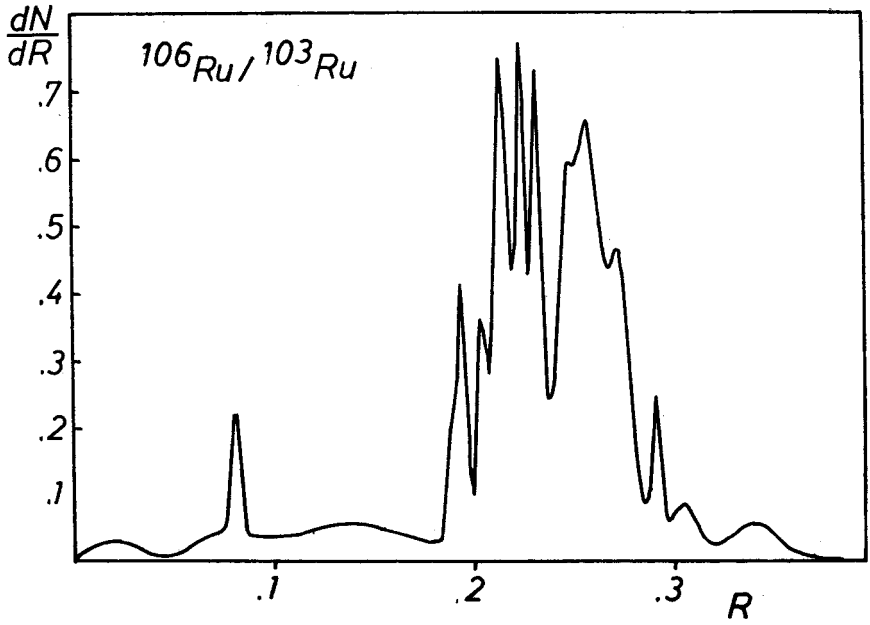

Fig. 8. Distribution of the $^{106}Ru/^{103}Ru$ isotope ratio in analysed particles

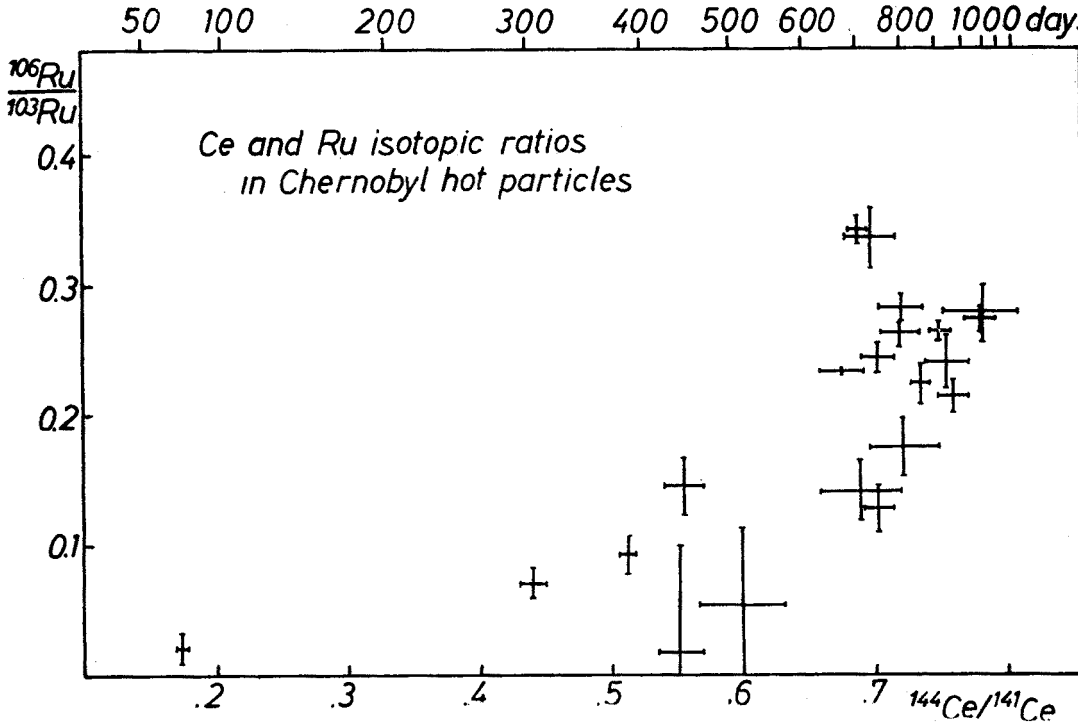

Fig. 9. Correlation of cerium and ruthenium isotope ratios established in analysed particles (see text)

For 17 hot particles both cerium and ruthenium isotopic ratios could be determined and Fig. 9 shows correlation between them. Generally the expected correlation is observed indicating that both, cerium and ruthenium present in a specific particle originated from the same fuel element. However, one easily notices that the correlation is not unique and the only reasonable explanation is a different fuel composition in various parts of the reactor core. In principle, for each of 17 pairs of ratio values a similar analysis can be made as shown in Fig. 5, nevertheless looking at coreesponding values e.g. for particles HS12 and HS16 of Table IV it is immediately apparent that they originated from parts of the fuel with drastically different composition.

Specifically the analysis gives 800 days with 63% of ^{239}Pu fission events in the first case and 620 days with 12% in the second one.

4.2. Activity ratio for radionuclides of various elements

Inspection of hot particles with respect to relative abundances of radioisotopes of various elements may yield information on physical and chemical processes taking place during the accident. All of the present work analysis assumes steady reactor work, but to minimize effects from possible variation of the reactor power the isotopes with similar half-lives were selected for comparison.

A regular appearance in gamma spectra of Ce, Nb and Zr lines, with remarkably stable relative intensities indicate similar behaviour of all three elements in processes which

led to hot particle formation. Particularly it is the case for Ce and Zr as seen from the distribution of the $^{141}Ce/^{95}Zr$ ratio shown in Fig. 10. Almost all values are narrowly scattered around the average value of 1.21, which is somewhat larger than 0.97 expected for this ratio saturation value assuming earlier deduced average fuel composition. A higher power level of the reactor for few months before an accident could lead to such an increase due to different half-lives of ^{141}Ce and ^{95}Zr, but such possibility has to be rejected since the corresponding correction would introduce further increase of already large isotopic

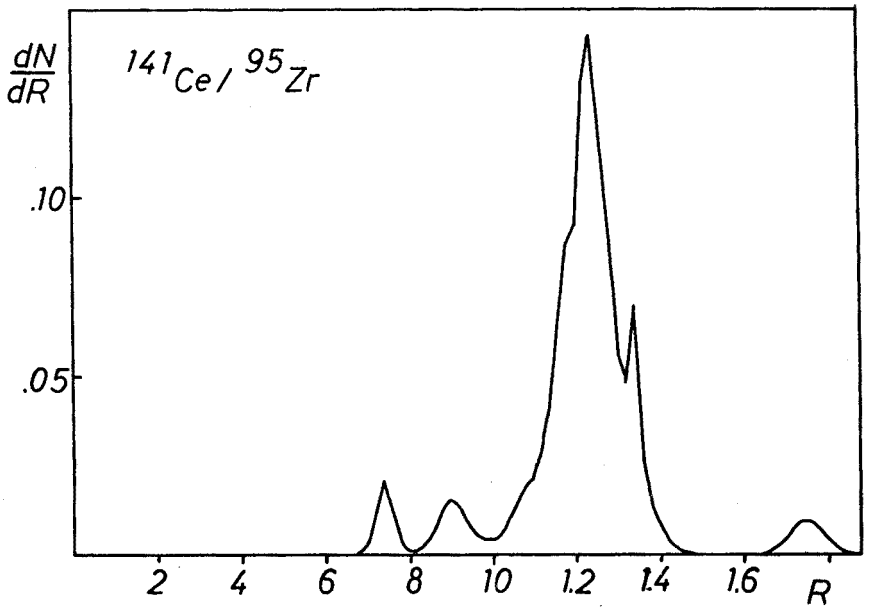

Fig. 10. Distribution of $^{141}Ce/^{95}Zr$ activity ratio observed in second type particles

ratios discussed above. Thus the observed value seems genuinely to indicate that in the hot particle formation process on average about 20% of zirconium as compared to cerium was in some way separated. A closer look at values listed in Table IV reveals small but significant differences, and in few cases the ratio values depart visibly from the average. Whereas high value for particle HS2 is trivially related to incomplete saturation (approx. 2.5 months old fuel element), small values observed for particles HS6 and M30 indicate that more of zirconium was preserved in those particles. On the other hand an extraordinary particle M44 quoted earlier contains predominantly cerium activity with barely visible traces of zirconium radionuclides.

The $^{95}Nb/^{95}Zr$ ratio distribution is shown in Fig. 11. It is visibly broader than the previous one, but to some extent this is caused by larger uncertainties.

The ^{95}Nb is produced as a daughter activity in a decay of ^{95}Zr and for bulk part of the reactor core a complete equilibrium was reached demanding that both activities are equal to each other. At an instant of accident the production of ^{95}Zr stops and both zirconium

and niobium are subjected to various processes, in which an initial $^{95}Nb/^{95}Zr$ ratio is settled. A subsequent decay changes the activity ratio in time until, after approx. 500 days the equilibrium value of 2.2 is reached. Practically, after time longer than 250 days the measured ratio becomes insensitive to its starting value. From the measurements of the present work it was still possible to extract the initial ratio values, but uncertainties are rather large in spite of the fact that the measured ratios were very accurate. Yet, it is seen from Fig. 11 and numbers of Table IV that the distribution is spread towards values smaller than 1.0, which indicates that on average niobium escaped more effectively than zirconium in a hot

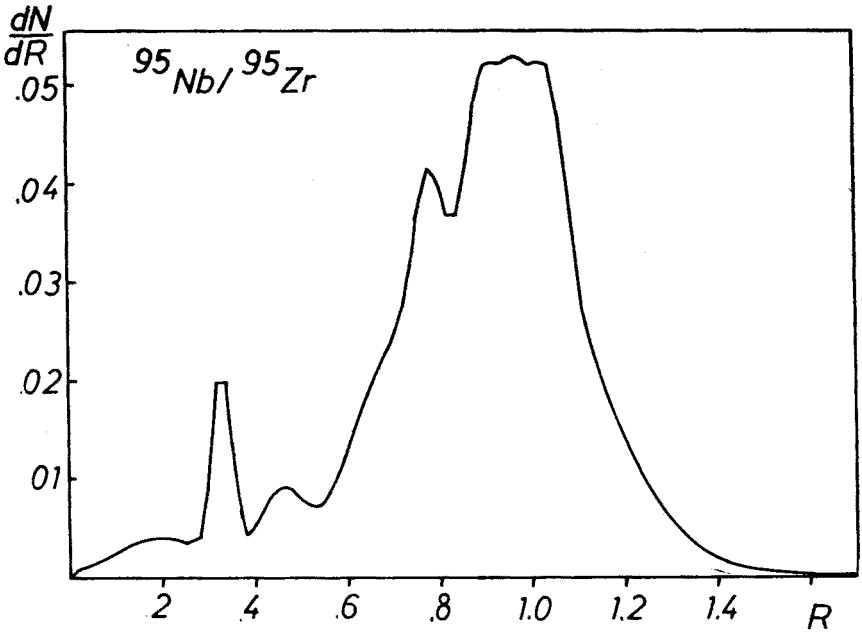

Fig. 11. Distribution of initial $^{95}Nb/^{95}Zr$ activity ratio observed in second type particles

particle formation process. Extraordinary particle M39 (see Fig. 4) is an example of niobium, which together with ruthenium was effectively separated from non-volatile cerium and zirconium.

The ruthenium radionuclides, as seen from Table IV, behaved in completely different way than other elements discussed above. The expected $^{141}Ce/^{103}Ru$ value of 1.5 was observed in few cases only; generally it assumes any value between zero and infinity. For number of particles ruthenium activity was reduced beyond the detection level. On the other hand 37 investigated species contained highly pure ruthenium activity and the question arises as to possible mechanism of a process which led to such effective separation. In a somewhat speculative way one can think of a possible scenario in which with access of oxygen a ruthenium volatile oxide was formed and gradually evaporated together with other volatile products. Most of it escaped directly to the atmosphere, but some part penetrated

hot, burning graphite moderator, where the deoxidation could happen. If the burning graphite temperature was above melting (2310°C) but below boiling (3900°C) point it could lead to the formation of ruthenium metal drops freezed inside graphite fragments ejected into atmosphere.

The 12.7 day activity of ^{140}Ba-^{140}La visible in some particles measured in July series yielded values, which point at barium as one of sizable constituents of second type particles. The amount of barium is varying from undetectable (in July) level to very high value as observed for Studsvik particle [5], but the data are to oscarce to extract more conclusive information. Future analysis of strontium radioisotopes in hot particles may provide better insight into behaviour of alkaline elements and the alpha spectroscopy analysis should shed a light on situation with actinides.

Small, but detectable amount of volatile products have been preserved in some particles. In Studsvik examples [5] the ^{131}J and ^{132}Te activities were seen, and the present analysis established a presence of cesium radionuclides. It should be noted that the ^{134}Cs/^{137}Cs isotopic ratio determined in few cases was clearly different from the average value of 0.53 determined in the radioactive fallout [2]. This indicates that cesium originated from the same fuel element as the rest of the particle and consequently the evaporation process was not exceedingly violent.

4.3. Final remarks

The gamma analysis of hot particles described in the present work provided information concerning some details of the Chernobyl power reactor and gave insight into general features of processes taking place during accident.

From the observed cerium and ruthenium isotopic ratios it is concluded that the fuel composition was different in various parts of the reactor core, which on average was for 570 days in operation. The distribution of radioisotopes of various elements gave information on behaviour of elements with different volatility properties in time of the accident. The possible mechanism explaining the observed separation of ruthenium particles was suggested.

The obtained results provide a basis for future analysis of alpha emitters [6] and strontium radionuclides contained in the Chernobyl fallout.

The estimate of the health risk connected with hot particles presence in a fallout was not a merit of the present work, but remarks given in Ref. [4] seem to be appropriate. Of particular concern is a possibility of particles being inhaled and the limitation of outdoor activities in first days after the accident was essential in minimizing possible consequences.

The author is grateful to A. Z. Hrynkiewicz, K. Królas, E. Obryk and several participants of the International Mikołajki School on Physics for many useful discussions. A consent given by the author's collaborators, Z. Szeglowski and K. Zuber, to quote some information prior to publication is greatly appreciated. Warm thanks are due to J. Grębosz whose technical assistance in preparing the data for publication has been extremely helpful.

REFERENCES

[1] Quoted in B. Goss Levi article, *Physics Today*, July 1986, p. 17.
[2] R. Broda, J. Grębosz, J. Wrzesiński, Raport No 1330/B, June 1986, Institute of Nuclear Physics, Kraków.
[3] K. Królas, to be published.
[4] C. Hohenemser, M. Deicher, A. Ernst, H. Hofsäss, G. Lindner, E. Recknagel, *Environment*, Vol. 28, No 5, June 1986.
[5] L. Devell, H. Todedal, U. Bergström, A. Appelgren, J. Chyssler, L. Andersson, *Nature* **321**, 192 (1986).
[6] R. Broda, Z. Szeglowski, K. Zuber, to be published.
[7] T. Goworek, private communication.
[8] C. M. Lederer, V. S. Shirley, *Table of Isotopes*, John Wiley and Sons, Inc., New York 1978.
[9] B. F. Rider, M. E. Meek, *Compilation of Fission Product Yields*, NEDO 12154-2D, July 1977.