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Measurement of the $^{129}I/^{131}I$ Ratio in Chernobyl Fallout

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Abstract

Rainwater collected in the Munich area approximately one week after the Chernobyl reactor accident was investigated for its content of the radioisotopes ^{129}I ($T_{1/2}$ = 1.6 x 10^7 yr) and ^{131}I ($T_{1/2}$ = 8.04 d). For the time of release, an isotopic ratio of $^{129}I/^{131}I$ = 19 ± 5 was found. This value was obtained from a gamma-ray activity measurement of ^{131}I with a Ge detector and a concentration measurement of ^{129}I with accelerator mass spectrometry. From the measured ratio an operating time of the reactor prior to the accident in the vicinity of two years can be estimated, which is in fair agreement with estimates from other long-lived to short-lived radioisotope ratios in the Chernobyl fallout. Some measurements of ^{131}I activity in thyroids of persons living in the Munich area is also reported.

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1. Measurements of ^{131}I activity in human thyroids

In the few weeks following the Chernobyl reactor accident of 26 April 1986, one of the strongest activities in the fallout was ^{131}I ($T_{1/2}$ = 8.04 d). In particular, the 364 keV gamma radiation emitted in 81% of the ^{131}I decays, could easily be detected with a Ge detector. Since such a detector is a standard piece of equipment in almost every nuclear physics laboratory, we could readily identify the fallout when it came first down in Munich on 30 April 1986. Besides measuring ^{131}I in various environmental samples, such as water, grass, soil, milk, etc., we were also interested in the uptake of ^{131}I into the human thyroid. About 30% of all iodine inhaled or ingested ends up in the thyroid, depending somewhat on the functional condition of this gland. We could get resonably quantitative measurements by sitting ten to fifteen minutes in front of a Ge detector.

Table 1 summarizes measurements on six different persons living in the general Munich area. It shows that people living on the country incorporated about twice as much activity as compared to those living in the city. This is supposedly due to the faster removal of wet (and probably dry) precipitation in the city. The highest activity was measured for person #1 living on the country and drinking milk from cows fed with fresh grass. In the fourth column of Table 1 the activity has been converted into an integrated thyroid dose using a converseion of 0.2 mrem/Bq, which assumes a weight of 20 g for the thyroid of an adult person. Small children may have thyroids which are a factor of ten smaller, resulting in doses correspondingly higher. Higher ^{131}I activities in the range from 1000 to 7000 Bq (still not posing a health hazard) were measured [1] in thyroids of students returning to England from Minsk and Kiew. It is not difficult to imagine that people who lived close to the reactor must have incorporated orders of magnitude more ^{131}I activity.

2. Measuarement of the $^{129}I/^{131}I$ ratio

Although ^{131}I was the most obvious activity of iodine, several other radioisotopes of iodine are produced in the fission of ^{235}U and ^{239}Pu. Some of them such as ^{132}I and ^{133}I were also readily detected with Ge detectors. Despite of a short half-life of only 2.28 h, ^{132}I could be observed several days after the accident, because its longer-lived precurser ^{132}Te (78.2 h) was also released.

Table 2 lists all fissiogenic radioisotopes of iodine with half-lives longer than about one hour. By far the longest-lived isotope is ^{129}I with a half-life of 16 million years. Such a long-lived isotope will accumulate continuously in the fuel rods of the reactor. The much shorter-lived isotopes will reach equilibrium between production and decay in an operating time of the reactor corresponding to about three half-lives. If there is at least one isotope which has not yet reached equilibrium it is possible to estimate the operating time of the reactor by measuring the isotopic abundance of this isotope relative to one of the shorter-lived ones. This is possible because isotopic ratios remain essentially unchanged by the complex processes governing the transport of fission products from the fuel rods to the usually very remote places of observation.

Neglecting the burn-up of fissionable material and fission products, and the breeding of new fission material (e.g. ^{239}Pu), we can obtain a crude estimate of the operating time of a reactor from the relative

$$N_1/N_2 = [Y_1\tau_1(1 - e^{-t/\tau_1}]/[Y_2\tau_2(1 - e^{-t/\tau_2})] \qquad (1)$$

Here, N_1/N_2 is the isotopic ratio of two isotopes labeled 1 and 2, Y_1 and Y_2 are the fission yields, and $_1$ and $_2$ are the mean lives, respectively.

A pair of isotopes were this method can be applied is ^{129}I and ^{131}I. From the two, ^{129}I is much more difficult to measure at low abundances. In a sample of rainwater collected near Munich we measured on 6 May 1986 a ^{131}I activity of 560 ± 80 Bq/liter. Assuming ten times more atoms of ^{129}I than ^{131}I present in one liter of this water, we would still only getting one decay of a ^{129}I atom in ten days. This makes direct decay counting not practical. However, an analysis of such low levels of ^{129}I is possible through neutron activation, a method developed more than twenty years ago [3,4]. We have chosen to use another technique of very high sensitivity, accelerator mass spectrometry (AMS), where ^{129}I atoms are counted at high energy after an accelerator. This method has been shown [5,6] to be useable at very low ^{129}I concentrations in water. The measurements reported here were performed with the AMS facility [7] of the pelletron tandem accelerator at the Weizmann Institute of Science in Rehovot, Israel. In figure 1 a schematic lay-out of the AMS facility is shown. Here, only a very brief description of the measurement will be presented. A more detailed account of the measurement will be given elsewhere [8]. In order to measure the ^{129}I concentration in the rainwater of Munich, 6.1 mg of stable iodine (^{127}I) were added as carrier to 1.14 liter of water. Iodine was extracted by precipitation of AgI, which was then used as sample material in the sputter source of the tandem accelerator. Ions of ^{127}I and ^{129}I were accelerated alternately to about 140 MeV energy and measured as beam current (~1 particle-nA) in a Faraday cup or counted in a time-of-flight + ΔE-E detector system (~5 counts/sec), respectively.

From these measurements a $^{129}I/^{127}I$ ratio of $(1.0 \pm 0.2) \times 10^{-9}$ was found in the rain water, whereas a measurement with a blank (distilled) water sample spiked with the same stable iodine material resulted in a ratio of (1.0 ± 0.4)

x 10^{-12}. It is worth mentioning, that these ratios were determined in an absolute sense, that is from a direct comparison of the beam current (^{127}I) and the ion counting rate (^{129}I). The uncertainty arising from this way of measuring an isotopic ratio is included in the quoted errors, contributing in particular to the total error of the rain water sample. The error of the blank sample is dominated by counting statistics. From the $^{129}I/^{127}I$ ratio in the rain water sample one obtains a ^{129}I concentration of (2.5 ± 0.5) x 10^{10} atoms/liter. Together with the ^{131}I activity we then calculate for the time of release (assumed to be 26 April 1986) a $^{129}I/^{131}I$ ratio of 19 ± 5.

3. Comparison of the $^{129}I/^{131}I$ ratio with other pairs of radioisotopes

In addition to the isotopes measured in this experiment, there were other pairs of radioisotopes in the Chernobyl fallout which can be used to estimate the reactor operating time. In Table 3 relevant parameters of these isotopes are listed. Various operating times using equation (1) and measured isotope ratios are summarized in Table 4. The values for the isotopic ratios of $^{90}Sr/^{89}Sr$ and $^{106}Ru/^{103}Ru$ references. No errors were reported in these measurements. An estimate of the operating time from equation (1) is complicated by the fact that the fission yield ratios, Y_1/Y_2, depend on whether they originate from fission of ^{235}U or ^{239}Pu (see Table 3). Since the ratio of ^{235}U to ^{239}Pu in the fuel material is not known the operating time given in the last two columns of Table 4 is calculated for both pure ^{235}U and pure ^{239}U fission.

Most sensitive to the ^{239}Pu content of the fuel is the $^{106}Ru/^{103}Ru$ ratio, where the measured isotopic ratio cannot be reproduced with the assumption of ^{235}U fission only. In principle, the $^{90}Sr/^{89}Sr$ ratio would be most suitable for the calculation of an operating time, since the fission yield ratios of

these isotopes are almost identical for ^{235}U and ^{239}Pu (compare Table 3). This means that the operating time estimated from the isotopic ratio is nearly independent of the fuel composition. Unfortunately, the activity of these Sr isotopes is difficult to measure, which seems to be reflected in the large disparity of the two quoted measurements. Our result of the $^{129}I/^{131}I$ ratio gives an operating time in the vicinity of two years. The estimates from the other radioisotope pairs seem to be in fair agreement with this result.

The ^{131}I activity from the Chernobyl fallout has been measured in many places all over Europe and also elsewhere in the world. Together with our result of the $^{129}I/^{131}I$ ratio one can calculate the ^{129}I fallout in these places, which in some cases will lie well above the natural and otherwise produced anthropogenic ^{129}I level. Using sensitive detection techniques such as the one presented here this spike of ^{129}I may turn out to be a useful tracer for hydrological and other environmental studies in the future.

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Table 1. Activity of ^{131}I in human thyroids (Munich area).

Date of measurement	Person #	Activity[a] (Bq)	Main living site	Dose[b] (mrem)
6 May 1986	1	227	country	45
	2	98	city	20
	3	91	city	18
	4	108	city	22
11 May 1986	2	60	city	12
	5	120	country	24
	6	117	country	23
27 May 1986	2	15	city	3
	5	36	country	7

[a] 1 Bq - 1 decay/sec. Uncertainty = ± 20%.

[b] Converson = 0.2 mrem/Bq.

Table 2. Radioisotopes of iodine produced in the fission of ^{235}U and ^{239}Pu

Isotope	$T_{1/2}$	Fission Yield (%)[a]	
		^{235}U	^{239}Pu
^{129}I	1.6×10^7 yr	0.74	1.5
^{131}I	8.04 d	2.88	3.85
^{132}I	2.28 h	4.30	5.39
(^{132}Te	78.2 h	4.30	5.39)
^{133}I	20.8 h	6.70	6.98
^{134}I	50.2 m	7.8	7.6
^{135}I	6.59 h	6.5	7.6

[a] Total cumulative fission yields [2].

Table 3. Pairs of radioisotopes produced in fission, consisting of a longlived and shortlived isotope.

Isotope	$T_{1/2}$		Fission Yield (%)[a]	
			^{235}U	^{239}Pu
^{90}Sr	28.8	yr	5.91	2.11
^{89}Sr	50.5	d	4.88	1.71
^{103}Ru	368	d	0.402	4.3
(^{132}Te	39.4	d	3.04	6.9
^{129}I	1.6×10^7	yr	0.74	1.5
^{134}I	8.04	d	2.88	3.85

[a] Total cumulative fission yields [2].

Table 4. Reactor operating time calculated from isotopic ratios and fission yields with equation (1)

Isotope Ratio	Measured Ratio	Reference	Operating time (yr)	
			^{235}U	^{239}Pu
$^{90}Sr/^{89}Sr$	19	[9]	3.1	3.1
	11	[10]	1.8	1.8
$^{106}Ru/^{103}Ru$	2.4	[9]	n.s.[a)]	0.78
	3.4	[10]	n.s.	1.3
	2.5	[11]	n.s.	1.4
	3.5	[12]	n.s.	0.82
	2.1	[13]	n.s.	0.65
$^{129}I/^{131}I$	19 ± 5	present	2.3	1.5

a) n.s. = no solution, since the measured isotope ratio value is higher than the equilibrium value.

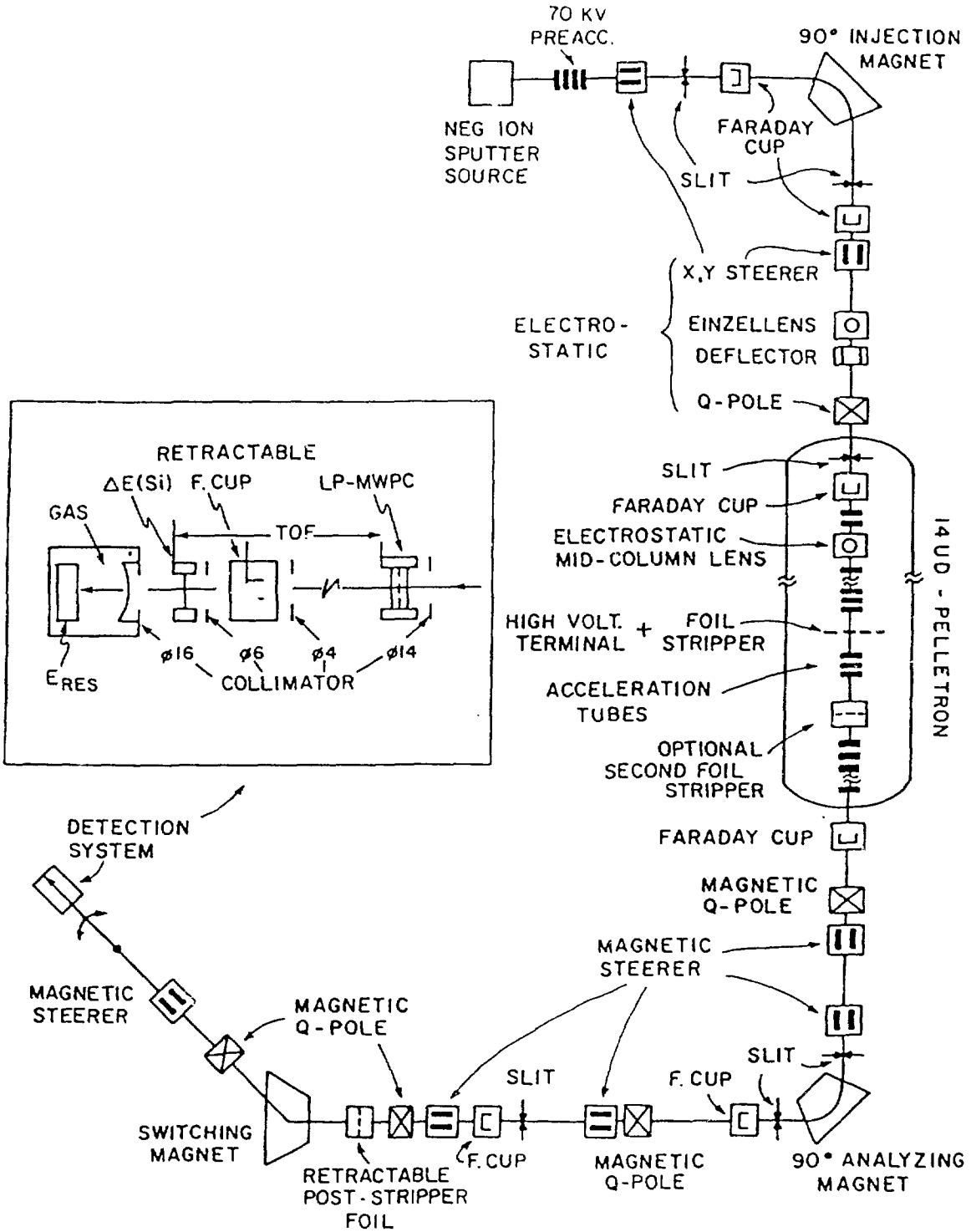

Fig.1 Schematic lay-out of the AMS system at the Weizmann Institute of Science in Rehovot, Israel.