NEUTRON-RICH NUCLIDES OBTAINED USING THE OSIRIS FACILITY

The OSIRIS Collaboration,

B. Grappengigsser, E. Lund and G. Rudstam,
Studsvik, Nyköping, Sweden.

Abstract

A survey of the short-lived nuclides produced in the isotope-separator-on-line facility at Studsvik is presented. Fission fragments recoiling out of a ^{235}U-target are caught by a graphite cloth in the ion source. The temperature of the ion source is kept high enough to release eighteen of the elements produced by fission so fast that it is possible to detect their short-lived isotopes after electromagnetic mass separation. No chemical separation but for the element-dependent separator efficiency has been used.

During the course of the work several methods for the determination of the half-life of the radioactive nuclides have been employed. The decays have been counted by means of the following systems: a $4\pi\beta$-counter made of plastic scintillating material, a neutron detector consisting of BF_3-tubes, an anthracene crystal with beta pulses sorted according to energy, and Ge(Li)- and Si(Li)-detectors for following the decay of gamma-rays, X-rays, and conversion electrons. The results are collected in a table containing half-life determinations of 75 nuclides or isomers, 32 of them new or with half-lives significantly different from published data.

1. INTRODUCTION

When the OSIRIS facility[1)] for mass separation of fission products came into operation in June 1968 it was considered of urgent interest to survey the nuclides obtained. This initiated a systematic investigation of the radioactive components appearing in the different mass chains. Various instrumentation for determining the half-life of the nuclides produced have been used. Results from this survey are presented in Table 1. As a target+ion source arrangement giving essen-

tially no element discrimination has been used in these experiments (see Ref.[1]) the element assignments given in the table are mostly based on a comparison with half-lives published in the literature and reasonable "parent-daughter" arguments. In a few cases the assignments have been confirmed by observing K-X-rays and conversion electrons.

2. INSTRUMENTATION

2.1 Integral counting of beta particles and of neutrons (Experiment I)

(B. Grapengiesser, E. Lund and G. Rudstam)

Most of the results in Table 1 were obtained with a $4\pi\beta$-detector connected to the magnetic tape transport system of the isotope separator. After a predetermined collection time the sample was rapidly moved to the detector for measurement, usually multiscaling, during a fixed period of time. This cycle was then repeated automatically in order to reach sufficient statistics. The sample stays all the time in the same vacuum system, and the time loss during the transportation of the sample between the collection position and the measuring position - a distance of about 1 meter - could be kept as small as 2 sec. This means that the practical limit for measurements lies in between 0.5 and 1 sec. depending on the strength of the samples produced.

The most important features required for the beta detector are the following:

i) a high efficiency which is constant, reproduceable and little dependent on the beta energy;

ii) a low background to make possible measurement even of very weak samples;

iii) capability of measuring high as well as low counting rates with precision.

A plastic scintillator was considered to best fulfill the third requirement. It was therefore chosen for the construction shown in Fig. 1, which well responds to the first two requirements. Its volume

is only 2.5 cm^3 which means easy shielding and a low background rate, typically 10 cps at full reactor power, and its solid angle is almost 4 π. The electronic components (see Fig. 1) accept high counting rates. Most of the results were obtained using a Nuclear Data Model 2200 analyzer in the multiscaling mode, but for more long-lived nuclides a printing scaler can be connected to the detector.

The discriminator level was set to accept pulses corresponding to electrons of energy larger than 50 - 200 keV. Although roughly half of the electrons have to pass through the tape (5.6 mg/cm^2) the efficiency was generally over 90 % in the measurements reported here.

The dead time was around 0.3 μsec in experiments without the prescaler. The fast prescaler arrangement shown in Fig. 1 should considerably decrease the dead time. Corrections for dead time losses have usually been negligible.

The possibility that certain elements diffuse out of the tape thereby causing the decay curve to appear too steep must be considered. This can be checked for long-lived components with known half-lives. No such effect was observed.

The multiscaling data are recorded on punched paper and evaluated by means of a computer programme which resolves the decay curve into a given number of components using an iterative procedure. Systematic trends in the differences between measured points and best fit reveal components not taken into account, and a new evaluation with better input data can be carried out. Obviously, two components with similar half-lives might not always be resolved by this method. In such cases a specific detection method, i.e. following the decay of a gamma line or X-ray line, would be preferable to the non-specific beta measurement.

The errors given for the $4\pi\beta$-data in Table 1 are purely statistical. Possible systematic errors caused by several components having closely the same half-life, or by contaminating activities of half-lives similar to that of the measured nuclide are not included.

The latter reason of systematic error can usually be ruled out for cases where duplicate measurements give consistent results as the degree of contamination and hence the effect would presumably vary from experiment to experiment. Cases with several determinations (the number of determinations is denoted in the table) generally show very good internal and external consistency indicating negligible effect of contaminating activities. Cases with only one determination should be considered as preliminary, however, until duplicate runs confirm the results.

For a few mass chains the decay was also followed using a neutron detector consisting of ten BF_3- tubes imbedded in paraffine and arranged in a semicircle around the sample position. The BF_3-tubes were coupled in parallel to a multiscaler for analysis of the decay of delayed neutron activities. The half-lives obtained in this way are also included in the table.

2.2 Anthracene detector for selective counting of beta particles (Experiment II)

(I. Andersson, S. Borg, L.-E. De Geer, G. Holm, S.G. Malmskog and B. Rydberg)

In order to be able to measure activities with very short half-lives (below one second) a selected ion beam is focused onto a second tape system where the detectors are arranged in such a way as to view the collection spot. Then the delay caused by the tape transport is avoided, and the moving tape is only used to remove long-lived activities. A special 1/2-inch tape with an aluminium coating was chosen because it can withstand the rather intense ion beams of stable krypton and xenon isotopes used as mass markers.

Both anthracene and Ge(Li)-detectors have been used to measure beta or gamma radiation. Usually the experimental sequence for the half-life determinations was supervised by a 8K PDP-9 computer. Such a sequence consisted of an accumulation on the tape for a predetermined time after which the ion beam was interrupted and the measurement was started. When using an anthracene detector, the output pulses were divided into four ranges, each corresponding to a 2 MeV energy interval. This enhances

short-lived activities with large Q-values in the higher energy gates. The PDP-9, run in a multiscaler mode, was used for storing the pulses from the different gates simultaneously. A similar procedure was adopted in some confirming experiments using the Ge(Li)-detector, now with gates set on different gamma lines.

All the data were analyzed on the PDP-9 using various fitting programmes. The resulting half-lives (averages of measurements using different energy gates) are given in Table 1.

2.3 Multispectrum scaling of gamma-ray, X-ray and conversion electron spectra from Ge(Li)- and Si(Li)-detectors (Experiment III)

(B. Fogelberg, A. Bäcklin and G. Hedin)

Identification of several short-lived nuclides have been made in the mass-region A = 114 - 125 by multispectrum scaling experiments in which the tape transport system mentioned in subsection 2.2 has been used. Ge(Li)-detectors for gamma-ray measurements and high resolution Si(Li)-detectors for X-ray or conversion electron measurements were used. For background reasons the detectors were in some cases placed 11 cm away from the beam implying a time delay of 0.1 sec for the movement of the tape before the measurements could be started. After an appropriate collection time the beam was shut off, and successive spectra were recorded in a ND 3300 multichannel analyzer operated in the 4 x 1024 channel mode. Generally, one spectrum was measured during the collection time, and three quadrants of the analyzer memory were used to record the successive decays. The cycle was automatically repeated until sufficient statistical accuracy was reached. Half-lives determined in this way are not particularly accurate since only three points on the decay curves are obtained, but reliable identifications of elements could be made from X-ray energies and from the K to L line distances in the conversion electron spectra. In some cases elements could also be identified through the presence of gamma transitions known to take place in the daughter nuclei.

The results obtained (averages of measurements using different gamma gates) are included in Table 1.

3. EXPERIMENTAL RESULTS

The experimental results are presented in the table below

Table 1

Half-lives of nuclides produced by OSIRIS

Mass number	Half-life	Experiment	Gamma gate, MeV	Element suggested	Comment	Literature Half life	Assignment	Ref
75	9±2 s	II		Zn	Ga X-rays observed			
76	5.4±0.3 s	I(1)[a]		Zn				
76	6±1 s	II		Zn	Ga X-rays observed			
76	29.0±0.3	I(1)		Ga		32 s	Ga	2)
77	1.4±0.3 s	II		Zn				
77	12.7±0.2 s	I(1)		Ga				
77	54.0±0.6 s	I(1)		^{m}Ge		54 s	^{m}Ge	2)
78	4.9±0.2 s	II		Ga				
79	2.86±0.04 s	I(1)		Ga, Ge				
79	18.5±0.2 s	I(1)		Ge				
79	8.2±0.2 m	I(1)		As		9.0 m	As	2)
80	1.7±0.2 s	II		Ga				
80	24±1 s	II		Ge				
81	43±1 s	I(1)		As		33 s	As	2)
82	16±1 s	II		As				
83	14±1 s	II		As		14.1 s	As	3)
83	110±10 s	II		As, Se		70 s	Se	2)
85	18.8±1.6 s	I(1)		Se				
85	2.87±0.01 m	I(1)		Br		3.00 m	Br	2)
87	56.0±0.3 s	I(1)		Br	Neutrons counted	55.6 s	Br	2)
88	16.6±0.4 s	I(1)		Br		16.3 s	Br	2)
89	4.6±0.3 s	II		Br		4.5 s	Br	2)
89	4.55±0.10 s	I(1)		Br	Neutrons counted			
91	9.02±0.09 s	I(1)		Kr		8.57 s	Kr	4)
91	59.2±0.2 s	I(1)		Rb		58.2 s	Rb	4)

Mass number	Half-life	Experiment	Gamma gate, MeV	Element suggested	Comment	Literature Half life	Literature Assignment	Ref
92	4.56±0.02 s	I(1)		Rb		4.50 s	Rb	4)
113	65.7±0.8 s	I(1)		Ag		1.2 m	Ag	2)
114	≤10 s	III	0.558	Ag	Ag and Cd X-rays observed. 0.558 MeV γ takes place in Cd	4.5 s	Ag	2)
115	17±5 s	III	0.131 0.229 0.389	Ag	Cd X-rays observed	49 s	Ag	5)
115	~ 20 m	III	0.227 0.229	Ag		20.0 m	Ag	2)
116	8.7±0.2 s	I(1)		Ag				
116	11±3 s	III	0.513 0.705	Ag	Cd X-rays observed. 0.513 MeV γ converts in Cd			
116	2.68±0.01 m	I(1)		Ag		2.5 m	Ag	2)
116	3±1 m	III	0.513 0.700 1.213	Ag	Weak Ag X-rays observed. Strong Cd X-rays observed			
117	5.34±0.06 s	I(2)		Ag				
117	73.1±0.4 s	I(2)		Ag		66 s	Ag	2)
117	80±10 s	III	0.135 0.311 0.337	Ag	0.135 MeV γ takes place in Cd			
118	4.00 ± 0.02 s	I(2)		Ag				
118	4.5±1 s	III	0.487 0.677 I.T. 0.129	^{m}Ag	Strong Ag X-rays observed. Weak Cd X-rays observed. 0.129 MeV γ converts in Ag, 0.487 γ in Cd			
119	6.0±0.5 s	I(2)		Ag				
119	3.3±0.1 m	I(2)		Cd, In		2.7 m	^{m}Cd	6)
119	3.3±1 m	III	0.293 0.343 0.763 1.317	Cd	In X-rays observed	2.8 m	In	6)
120	1.3±0.3 s	III	0.203 0.506 0.698	Ag	0.203 and 0.506 MeV γ converts in Cd Ag and Cd X-rays observed			
120	2.87±0.05 s	I(2)		In		3.2 s	In	2)
120	~ 5 s	III	1.171	In	1.171 Mev γ takes place in Sn			

Mass number	Half-life	Experiment	Gamma gate,MeV	Element suggested	Comment	Literature Half-life	Assignment	Ref
120	50.8±0.5 s	I(2)		In		46 s	In	2)
120	~ 60 s	III	0.1946 1.171	In	0.1946 MeV γ converts in Sn. Sn X-rays observed			
121	≲ ~ 3 s	III	0.194 0.420 0.448	Ag	Cd X-rays observed			
121	12±5 s	III	0.334 0.349 1.039	Cd	In X-rays observed	12.8 s	Cd	2)
121	25±3 s	III	0.261 0.657 0.925	In	Sn X-rays observed	30 s	In	2)
121	3.76±0.06 m	I(1)		In		3.1 m	In	2)
121	~ 3 m	III	0.0609	In	Sn X-rays observed			
122	5.5±0.1 s	I(1)		Cd, In				
122	~ 5 s	III	X-ray	Cd[b)]	In X-rays observed.			
122	12.6±0.3 s	I(1)		In		8 s	In	2)
122	9±2 s	III	0.1031	In	Sn X-rays observed. 0.1031 MeV γ converts in Sn			
123	5.97±0.05 s	I(3)		Cd, In		10 s	In	2)
123	47.8±0.5 s	I(2)		In		36 s	In	2)
123	38.9±0.4 m	I(1)		Sn		40.1 m	^{m}Sn	7)
124	3.21±0.06 s	I(2)		In		3.6 s	In	2)
124	3.5±1 s	III	1.132	In	1.132 MeV takes place in Sn			
125	2.32±0.03 s	I(2)		Cd, In				
125	3.5±1 s	III	1.333	Cd, In	Weak In X-rays observed			
125	12.2±0.2 s	I(2)		Cd, In				
125	7.78±0.07 m	I(1)		Sn		9.6 m	^{m}Sn	7)

Mass number	Half-life	Experiment	Gamma gate,MeV	Element suggested	Comment	Literature Half-life	Assignment	Ref
126	1.53±0.01 s	I(3)		In				
127	3.64±0.04 s	I(2)		In				
127	2.0±0.4 s	II		In				
127	4.4±0.1 m	I(2)		Sn		4.1 m	Sn	2)
128	3.7±0.5 s	I(2)		In				
129	0.8±0.3 s	II		In				
130	0.5±0.2 s	II		In				
130	6.5±0.1 m	I(1)		Sn, Sb		7.1 m	Sb	2)
131	0.3±0.1 s	II		In				
133	1.7±0.3 s	II		Sn				
133	9±2 s	II		^{m}I				
135	19.2±0.1 s	I(2)		Te		18 s	Te	8)
135	18±4 s	II		Te				
137	24.6±0.2 s	I(1)		I		24.4 s	I	2)
137	24.5±0.2 s	I(1)		I	Neutrons counted			
137	4.00±0.05 m	I(2)		Xe		3.82 m	Xe	4)
138	6.57±0.12 s	I(3)		I		5.9 s	I	2)
138	6.8±0.3 s	I(1)		I	Neutrons counted			
139	2.46±0.15 s	I(2)		I	Neutrons counted	2.7 s	I	2)
139	39.3±0.9 s	I(2)		Xe		39.68 s	Xe	4)
139	9.53±0.10 m	I(2)		Cs		9.27 m	Cs	4)
141	25.6±0.6 s	I(1)		Cs		24.7 s	Cs	4)
143	12±1 s	I(1)		Ba		12 s	Ba	2)
145	6.6±0.5 s	I(1)		Ba		5.6 s	Ba	9)
145	36.6±0.4 s	I(1)		La		29.2 s	La	9)
145	4.43±0.10 m	I(1)		Ce		3.0 m	Ce	2)
146	38±9 s	I(1)		La, Ce				
	19±2 m	I(2)		Ce, Pr		14 m 24 m	Ce Pr	2)
147	13±3 s	I(1)		La, Ce		65 s	Ce	2)

a) Figure within brackets denotes the number of determinations.

b) Indium X-rays show growth corresponding to a ~ 5 sec. parent activity. This is more probably due to the decay of cadmium than of a very low-energy indium isomer.

REFERENCES

1) S. Borg, I. Bergström, G.B. Holm, B. Rydberg, L.E. De Geer, G. Rudstam, B. Grapengiesser, E. Lund and L. Westgaard, On-line Separation of Isotopes at a Reactor in Studsvik (OSIRIS), to be published in Nucl. Instr. Methods.

2) C.M. Lederer, J.M. Hollander and I. Perlman, Table of Isotopes, Sixth Edition, John Wiley and Sons, Inc., New York 1967.

3) P. del Marmol, J. Inorg. Nucl. Chem. 30, 2873 (1968).

4) G.C. Carlson, W.C. Schick, Jr., W.L. Talbert, Jr. and F.K. Wohn, Nucl. Phys. A125, 267 (1969).

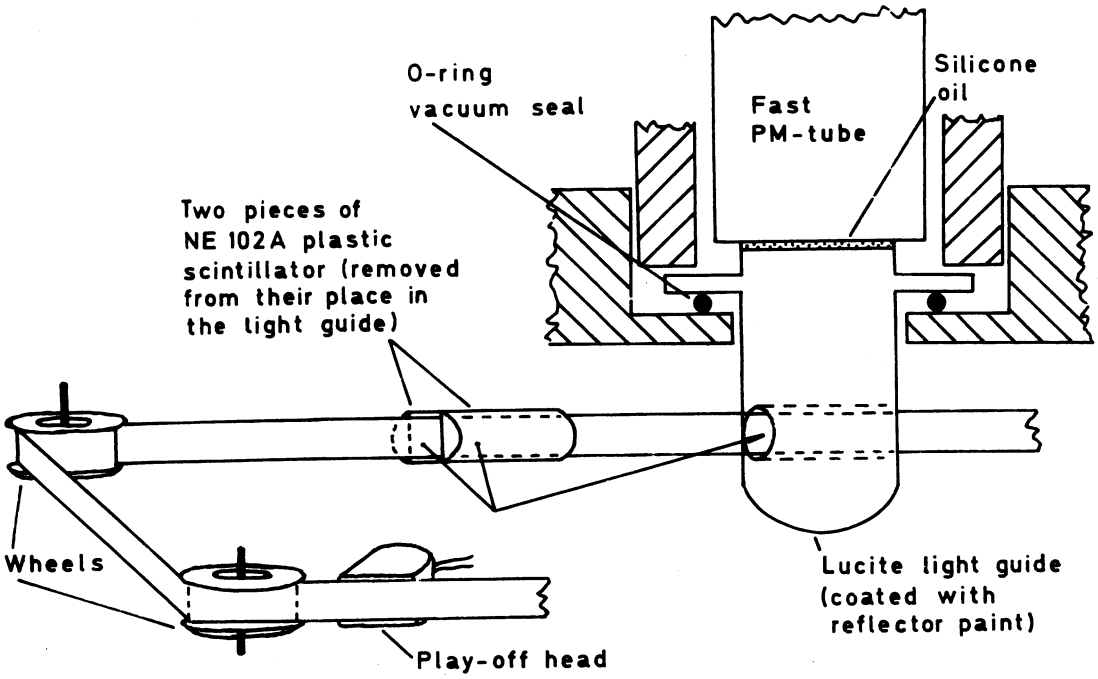
5) A. Kjelberg, A.C. Pappas and T. Tunaal, J. Inorg. Nucl. Chem. 30, 737 (1968).

6) E. Schwarzbach and H. Münzel, Radiochimica Acta 10, 20 (1968).

7) B.R. Erdal and A.C. Wahl, J. Inorg. Nucl. Chem. 30, 1985 (1968)

8) H.O. Denschlag, J. Inorg. Nucl. Chem. 31, 1873 (1969).

9) J.B. Wilhelmy, Thesis, Lawrence Radiation Laboratory Report UCRL-18978 (1969).



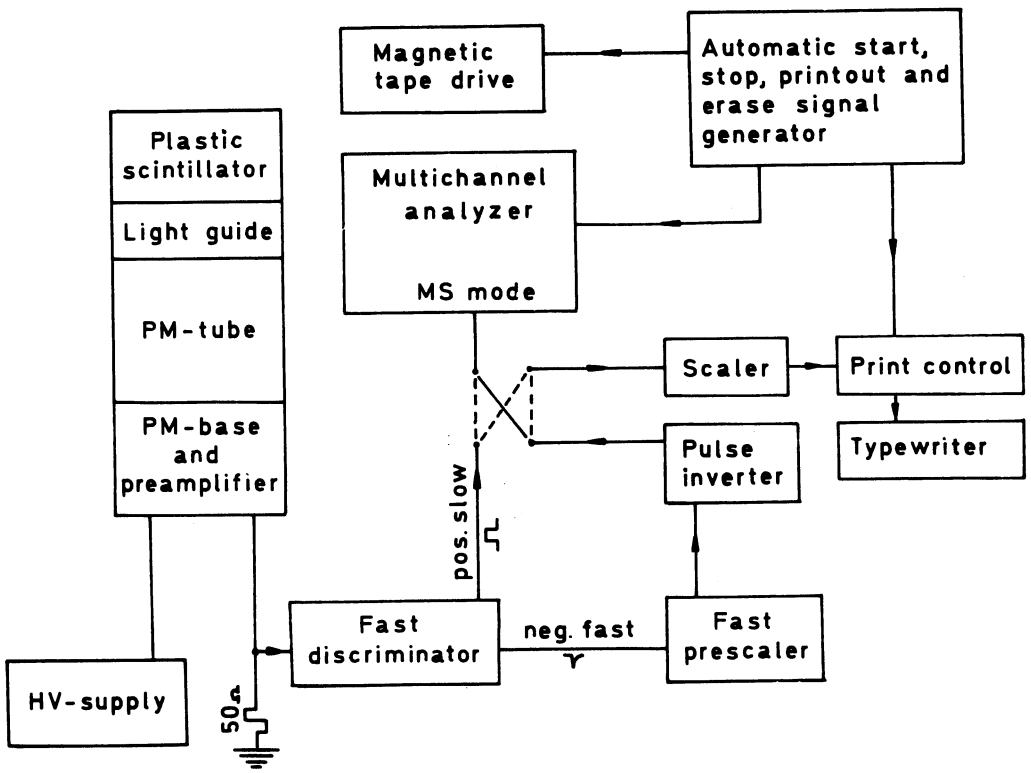


Fig. 1 4π beta plastic scintillator
detector arrangement.