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Fission Branch in (259)Lr and Confirmation of (258)Lr and (259)Lr Mass Assignments

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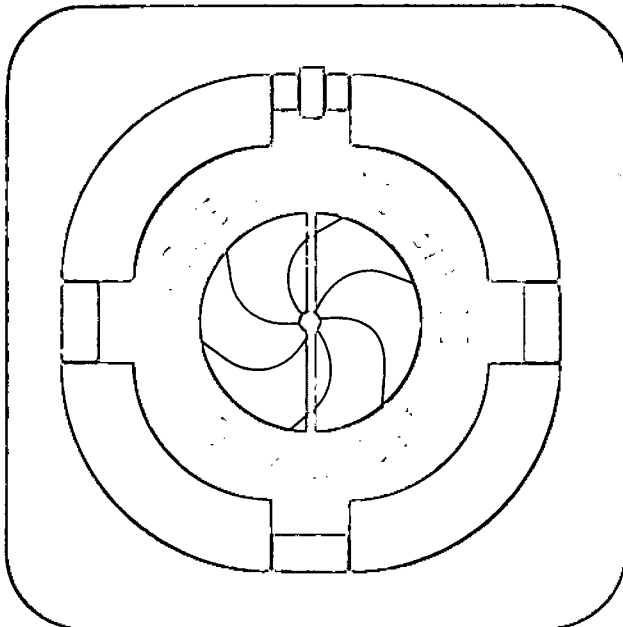
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Fission Branch in ^{259}Lr and Confirmation of ^{258}Lr and ^{259}Lr Mass Assignments

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Fission Branch in ^{259}Lr and Confirmation of ^{258}Lr and ^{259}Lr Mass Assignments

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Abstract

The mass assignments for the activities previously assigned to ^{259}Lr and ^{258}Lr have been confirmed. The ^{259}Lr and ^{258}Lr were produced by the $^{248}\text{Cm}(^{15}\text{N},4n)$ and $^{248}\text{Cm}(^{15}\text{N},5n)$ reactions, respectively. The Lr α -activities were measured directly using our rotating wheel system. Mass assignments were made by an α -daughter recoil catcher technique in which the ^{255}Md and ^{254}Md daughters were caught on aluminum foils placed directly above the rotating wheel. The α -decay of the 20.07-h ^{255}Fm and the 3.24-h ^{254}Fm granddaughter activities in these aluminum foils were then measured and correlations with the decay of the Lr parent activities were made based on the distribution of the Fm activities among the Al foils, which corresponded to the half-lives of the parent Lr α -activities. We have redetermined the half-lives for ^{259}Lr and ^{258}Lr to be $6.35_{-0.42}^{+0.46}$ s and $3.93_{-0.31}^{+0.35}$ s, respectively. The spontaneous fission branch in ^{259}Lr was measured to be $23\pm 2\%$. The spontaneous fission mass distribution appears to be broadly symmetric and the average total kinetic energy is deduced to be 200 ± 10 MeV.

PACS Numbers: 23.60.+e 24.75.+i 27.90.+b

Introduction

The observation of ^{259}Lr and ^{258}Lr was first reported by Eskola et al.¹ in 1971. They were produced in ^{15}N bombardments of ^{248}Cm targets, and were found to decay by emission of α -particles with half-lives of 5.4 ± 0.8 s and 4.3 ± 0.5 s, respectively. The spontaneous fission (SF) and electron-capture (EC) branches in the decay of ^{258}Lr were found to be small^{1,2} (<5%). The SF branch in the decay of ^{259}Lr was not measured.^{1,3} The EC branch in the decay of ^{259}Lr should be very small, based on the expected Q-value³ of 1.98 MeV and EC systematics. The mass assignments for these Lr isotopes were based on their excitation functions in the $^{15}\text{N} + ^{248}\text{Cm}$ reactions, although the low-energy portion of these excitation functions¹ was not measured well.

The discovery of 34-s $^{262}105$ by Ghiorso et al.⁴ was made by observing the α - α correlations arising from the α -decay of $^{262}105$ followed closely in time by the α -decay of its ^{258}Lr daughter (see Fig. 1). This α - α correlation technique was also used in subsequent studies of the chemical properties and decay of this isotope.^{5,6} Several unsuccessful attempts^{1,7-9} to produce the new isotope, $^{263}105$ have relied principally on this α - α correlation technique to look for the α -decay of ^{259}Lr daughter atoms following the α -decay of ^{263}Ha (see Fig. 1). The long-lived $^{263}105$ atom reported by Gregorich et al.⁸ could not be reproduced in subsequent experiments by those authors.¹⁰

The failure of all these experiments^{4,7-9} to detect the decay of the new isotope, $^{263}105$, which should be produced in quantities as large as, or larger than, those for $^{262}105$, brings into question the mass assignments of these Lr activities (and therefore the mass assignment of $^{262}105$). It is possible that the 5.4-s 8.45-MeV α -activity is due to the

decay of ^{258}Lr (or an isomeric state in ^{258}Lr). It is also possible that the 4.3-s 8.56- to 8.65-MeV α -activity assigned to ^{258}Lr is actually due to the decay of ^{259}Lr (and the 34-s 8.45- to 8.67-MeV α -activity assigned to ^{262}Ha would, therefore, be due to the decay of 263105). It was, therefore, necessary to experimentally confirm the mass assignments of ^{259}Lr and ^{258}Lr , and to determine the fission branch in ^{259}Lr before searching further for the new isotope 263105 .

Experimental Techniques

^{259}Lr and ^{258}Lr were produced at the Lawrence Berkeley Laboratory 88-Inch Cyclotron via the $^{248}\text{Cm}(^{15}\text{N},x\text{n})^{263-x}\text{Lr}$ reaction where $x=4$ and 5 , respectively. The target consisted of 0.49 mg/cm^2 ^{248}Cm (97% isotopic purity) deposited in a 6-mm diameter circle on a 2.75-mg/cm^2 beryllium foil as the oxide (0.54 mg/cm^2 Cm_2O_3) by the molecular plating method.¹¹ The 93-MeV beam of $^{15}\text{N}^{4+}$ ions passed through a 1.8-mg/cm^2 HAVAR (Hamilton Precision Metals) vacuum window, 0.3-mg/cm^2 of N_2 cooling gas, and the 2.75-mg/cm^2 Be target backing before passing through the ^{248}Cm target material. The energy of the ^{15}N beam in the target material was calculated¹² to be 79.8-80.6 MeV. The average beam intensity during the experiments was approximately 4.5×10^{12} particles/s. Products of nuclear reactions which recoiled out of the target were stopped in helium gas at 1.3 bar which had been seeded with potassium chloride aerosols. The reaction products, after attaching to the aerosols, were swept out of the recoil chamber through a polyvinyl chloride capillary (1.2 mm i.d.). They were transported through the capillary for a distance of 5 m to our rotating wheel system, the MG.^{13,14} At the MG, the activity laden aerosols were collected on $40\text{ }\mu\text{g/cm}^2$ polypropylene foils placed on the

periphery of an 80-position rotating wheel. The wheel was stepped at 5.4-s intervals so as to position the newly collected sample in the first detector station, while positioning the previous samples in the subsequent detector stations. Each of six detector stations held a passivated, ion implanted planar silicon (PIPS) detector (100 mm² active area) below the wheel to measure the energies and times of α -particles above 5 MeV and the energies and times of fission fragments, after they passed through the polypropylene foils. These detectors had an efficiency for the detection of α -particles of 30% and 60% for the detection of a SF fragment.

Aluminum foil (14 mg/cm²) were placed above the wheel at each of the detector stations. These aluminum foils subtended about 40% of 4π sr from the source positions on the wheel. When an α -particle was emitted from the source in a downward direction, the ≈ 150 keV recoil energy imparted to the daughter nucleus was sufficient to eject it from the source and imbed it in the top aluminum foil. At the end of an irradiation, these aluminum foils were removed and the radiations from the α -daughter recoils were measured in a second set of detectors. These "daughter detectors" consisted of ten 300-mm² surface barrier detectors which were set up to record the time and energy of any α -particles above 5 Mev and the time and energy of any fission fragments. Because of limitations imposed by the number of daughter detectors and other experiments using the detectors, only the first five of the aluminum collection foils from the six detector stations from each irradiation were counted. A schematic of the experimental counting and α -recoil catcher arrangement is presented in Fig. 2.

The decay of ²⁵⁸Lr and ²⁵⁹Lr and their daughter and granddaughter activities are shown schematically in Fig. 1. Three irradiations were

performed. During each irradiation, the α -recoil atoms were caught in fresh aluminum foils and α -particles and SF fragments were measured on-line with the bottom detectors at each of the detector stations. The first and third irradiations were 2.5 hours in duration, and the Fm granddaughter activities in the aluminum foils were measured for 14-21 hours. The second irradiation was 11.5 hours in length and the aluminum recoil catcher foils were counted for 114 hours. During all irradiations, the wheel containing the polypropylene foils was replaced every 36 minutes to prevent the buildup of long-lived activities in the on-line measurements.

Energy calibrations for all detectors were obtained by measuring the spectrum of α -particles from ^{212}Bi (6.051 and 6.090 MeV) and ^{212}Po (8.784 MeV) in equilibrium with a ^{212}Pb source. These α -peaks were used to calibrate each detector. On-line energy calibrations, using α -peaks present in the experimental spectra, were also used. Before summing spectra from the various irradiations, the data were normalized to the same energy calibration. Decay curves were produced by integrating the numbers of events in energy ranges containing the α -peaks of interest for appropriate sets of time intervals. All decay curve fits were performed with the Maximum Likelihood Decay by the Simplex method (MLDS) code.¹⁵ This maximum likelihood code is appropriate for analysis of the relatively small numbers of events observed in these experiments. Fission fragment energy calibrations for the on-line detectors were obtained by measuring ^{252}Cf sources through $40 \mu\text{g}/\text{cm}^2$ polypropylene foils.

Results

On-line measurements

The alpha-energy spectrum resulting from summing the data from all on-line measurements in all the irradiations is presented in Fig. 3. The main peak in the spectrum at 6.775 MeV is due to ^{213}Fr produced by the α -decay of ^{217}Ac which was in turn produced via the $^{208}\text{Pb}(^{15}\text{N},6n)$ reaction from a small lead impurity in the target. The 8.09-MeV peak is from ^{213}Rn produced by the 0.55% EC branch in the decay of ^{213}Fr . The ^{259}Lr α -peak at 8.45 MeV and the ^{258}Lr α -multiplet between 8.56 and 8.65 MeV are clearly visible. Fig. 4 shows single component decay curves for the SFs, and the ^{259}Lr and ^{258}Lr α -peaks from the on-line measurements. The most probable half-life for ^{259}Lr was found to be $6.35^{+0.35}_{-0.31}$ s. The stated error limits indicate the interval of equal likelihood chances¹⁶ corresponding to a confidence level of 68%, considering the covariance effects¹⁵ of the other free parameters. This value for the ^{259}Lr half-life is consistent (within the error limits) with the less precise previous value¹ of 5.4 ± 0.8 s. The most probable ^{258}Lr half-life was found to be $3.93^{+0.35}_{-0.31}$ s, again consistent with the less precise previously published values² of 4.35 ± 0.59 and 4.2 ± 0.6 s.

A total of 537 SF events was recorded during the irradiations. A 102-hour measurement of the background on the bottom detectors of the MG taken soon after the irradiations, showed a small fission background of 12 events. The decay curve presented in Fig. 4 has been corrected for this background contribution. When the SF decay was fit with a single component, the half-life was found to be $6.65^{+0.45}_{-0.39}$ s. This is consistent with the half-life value measured for the ^{259}Lr α -particles. When the half-life in the fit to the on-line SF data is fixed at the half-life of

6.35 s measured for the ^{259}Lr α -particles, the initial activities of the two curves indicate a ^{259}Lr SF branch of $23\pm 2\%$. The corresponding partial fission half-life of 27.6 s indicates a SF hindrance of 5.6×10^3 , which is consistent with odd-particle SF hindrance factors in the heavy actinide and transactinide region.¹⁷ Fig. 5 shows the SF single fragment kinetic-energy spectrum superimposed on a similar distribution for SF of ^{252}Cf . The most probable energies of the peaks in the ^{252}Cf single fragment energy spectrum were used to determine a linear energy calibration¹⁸ for fission fragments. The average fragment energies for ^{259}Lr and ^{252}Cf were multiplied by two to give estimates for their respective average total kinetic energies. An average post-neutron emission total kinetic energy for ^{259}Lr of 200 ± 10 MeV resulted after a small correction to these estimates was made to make the average post-neutron total kinetic energy for ^{252}Cf consistent with the recently reported¹⁸ value of 181.0 MeV. The ^{259}Lr average total kinetic energy is in the range of "normal" average total kinetic energies,¹⁹ and not the "high" average total kinetic energy indicating fission into very spherical, shell-stabilized fragments. The mass-yield distribution of the ^{259}Lr SF appears to be broadly symmetric.

It should be noted that the fission decay can also be fit with two components, a short component of 3.93 s (^{258}Lr) and a long component corresponding to 1.5-h ^{256}Md decaying to 2.6-h ^{256}Fm , which decays via SF. While it could be expected that ^{258}Lr has an EC branch to the 1.2-ms SF, ^{258}No , large EC or SF branches in ^{258}Lr were ruled out in the discovery experiment.¹ Large EC or SF branches in ^{258}Lr have also been ruled out in the radiochemical experiments^{5,6} performed on $^{262}105$, which α -decays to ^{258}Lr . In all of these experiments, 28 α - α correlations between $^{262}105$

and ^{258}Lr were recorded; in only one case was an α -SF correlation recorded, presumably from the α -decay of $^{262}\text{105}$ followed by the EC decay of ^{258}Lr to ^{258}No , a 1.2-s SF activity, indicating that the EC and SF branches account for less than a few percent of the total ^{258}Lr decay.

Off-line measurements

After each of the irradiations, the aluminum foils were removed from the tops of the detector stations in the MG and placed on a set of 300 mm² PIPS detectors to measure the 7.02-MeV ^{255}Fm and 7.19-MeV ^{254}Fm granddaughter α -activities produced by electron capture in the ^{255}Md and ^{254}Md daughters that recoiled into the aluminum foils from α -decay of the Lr isotopes. A summed spectrum of the measurements in the aluminum foils is presented in Fig. 6. The α -peaks due to the decay of 3.24-h ^{254}Fm (7.19 MeV) and 20.07-h ^{255}Fm (7.02 MeV) are clearly visible. The ^{255}Fm peak shows the high-energy tail characteristic of conversion electron summing, which provides proof that this peak is not due to the decay of ^{252}Fm (7.04 MeV). The other peaks in the spectrum are due to: 5.4-h ^{209}At (5.65 MeV), the α -decay daughter of 34.6-s ^{213}Fr , and 2.4-h ^{210}Rn (6.04 MeV), the α -decay daughter of 2.46-s ^{214}Ra . The peak at 6.26 MeV is due to background from the decay of 333.5-d ^{248}Cf which had recoiled into one of the detectors from the α -decay of ^{252}Fm in previous experiments (the broad peak shape is typical of α -activities which have recoiled into the detector face). The background peak at 6.63 MeV is due to the decay of ^{252}Es and ^{253}Es which also recoiled into one of the detectors from the α -decay of ^{256}Md and ^{257}Md in previous experiments.

Decay curves were constructed for each irradiation by taking the number of Fm daughter events detected in each of the five Al α -recoil catcher foils and assigning 5.4-s time intervals to each foil. These time intervals were 0-5.4 s for the foil above detector station 1, 5.4-10.8 s for the foil above detector station 2, and so on for the other foils. The distribution of Fm events among these foils is then indicative of the half-life of the Lr α -activity which caused the Md parent atoms of the Fm atoms to be imbedded in the 14 mg/cm² aluminum catcher foils. These decay data were fit with a single component. The half-lives were consistent with those observed for the ²⁵⁹Lr and ²⁵⁸Lr activities measured directly in the on-line measurements. Fig. 7 shows the decay curves, for half-lives fixed at the most probable values, as determined in the on-line measurements. From these decay curves, it is clear that the ²⁵⁵Fm is present in the aluminum catcher foils due to the recoil from a parent α -activity with a half-life consistent with that measured on-line for the 8.45-MeV activity measured on-line. Similarly, it is clear that the ²⁵⁴Fm is present in the aluminum foils due to the recoil from a parent α -activity with a half-life consistent with that measured on-line for the 8.56-8.65-MeV α -activity. The initial activities calculated from these fits were used to calculate the efficiency for catching daughter recoils from α -decay of the ²⁵⁹Lr and ²⁵⁸Lr. These efficiencies were 23±4%, consistent with the calculated solid angle subtended by the aluminum foils from the sources in the on-line measurements, after adjustment for self-adsorption of the recoils in the KCl of the gas-jet.

We also confirmed the identities of the ²⁵⁵Fm and ²⁵⁴Fm granddaughters of ²⁵⁹Lr and ²⁵⁸Lr by measuring the decay of the Fm activities with time. The decay of the ²⁵⁵Fm in the aluminum α -recoil

catcher was consistent with the known 20.07-h half life of ^{255}Fm . Fig. 8 shows this decay curve fit with the half-life fixed at 20.07 h. Similarly, the decay of the ^{254}Fm in the aluminum α -recoil catcher foils was consistent with the known 3.24 h half-life of ^{254}Fm . Fig. 9 shows this ^{254}Fm decay curve fit with the half-life fixed at 3.24 h.

Conclusions

- Assignment of an 8.45-MeV α -activity to the decay of ^{259}Lr has been confirmed by an α -recoil technique.
- The half-life of ^{259}Lr has been measured to be $6.35^{+0.46}_{-0.42}$ s from the α -decay of the 8.45 MeV peak observed in the on-line spectra..
- An SF activity with a half-life of $6.65^{+0.45}_{-0.39}$ s, consistent with that measured for the α -decay of ^{259}Lr , has been detected, and is therefore assigned to ^{259}Lr . It corresponds to a $23\pm 2\%$ SF branch in ^{259}Lr .
- The average total kinetic energy for SF of ^{259}Lr is estimated to be 200 ± 10 MeV based on single fragment measurements. The mass distribution is probably broadly symmetric, but kinetic-energy measurements of coincident SF fragments need to be made.
- Assignment of an 8.56 to 8.65-MeV α -activity to decay of ^{258}Lr has been confirmed by an α -recoil technique.
- The half-life of ^{258}Lr has been measured to be $3.93^{+0.35}_{-0.31}$ s.

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Figure Captions

Fig. 1. Schematic of the decay of ^{258}Lr and ^{259}Lr and their daughter and granddaughter activities as well as the parent activities $^{262}\text{105}$ and the undiscovered $^{263}\text{105}$. α -decay energies in MeV are indicated near the arrows signifying α -decay. Data are taken from previously published work^{1,3,5,6} except where differences were found in the current research (Lr half-lives and SF branch in ^{259}Lr).

Fig. 2. A schematic of the counting and α -recoil catcher setup (vertical scale exaggerated). The top half of the figure shows the setup in the MG chamber for the on-line portion of the experiment. Activities are delivered to the polypropylene foils via the gas-jet. The wheel is stepped at 5.4-s intervals to position the polypropylene foils in the six detector stations, each of which has a detector below the wheel and an Al catcher foil above the wheel. An event in which the α -particle from the decay of a Lr nucleus is detected in the detector of station 2 and the Md daughter recoils into the aluminum foil above the sample is depicted. After EC-decay of the Md daughter to Fm, the aluminum foils are removed and counted in the off-line portion of the experiment, shown in the bottom half of the figure. The α -decay of this Fm atom is depicted.

Fig. 3. The summed on-line α -particle spectrum taken in the detectors below the MG wheel. The continuum in the low-energy part of the spectrum is due to the decay of the $A=8$ isotopes produced by interactions of the beam with the beryllium target backing.

Fig. 4. The decay curves from the on-line measurements of the SFs, the ^{259}Lr α -peak at 8.45 MeV, and the ^{258}Lr α -peak at 8.56-8.65 MeV. In this figure, the average count rates during the time intervals are indicated by the symbols. For each fit, the center curve is the most probable fit to the data. The upper and lower curves are the limits which encompass 68% of the probability in a Poisson distribution centered on the number of counts expected during the interval from the most probable fit. These fits to the Lr α -peaks were used to determine the most probable half-lives for ^{259}Lr and ^{258}Lr .

Fig. 5. The single fragment kinetic-energy distribution for SF of ^{259}Lr superimposed on the single fragment kinetic-energy distribution for SF of ^{252}Cf .

Fig. 6. The summed α -particle spectrum from the off-line measurements of the aluminum α -recoil catchers.

Fig. 7. The decay curves from the distribution of the Fm atoms among the aluminum recoil catcher foils with half-lives fixed at values determined from the fits shown in Fig. 4. These decay curves are indicative of the half-lives of the Lr α -activities which caused the recoils to be implanted in the foils. The significance of the curves for each fit are the same as in Fig. 4.

Fig. 8. The decay curve for the ^{255}Fm in the aluminum α -recoil catcher foils from the long irradiation. The half-life has been fixed at 20.07 h. The significance of the curves are the same as in Fig. 4.

Fig. 9. The decay curve for the ^{254}Fm in the aluminum α -recoil catcher foils from the three irradiations. The half-life has been fixed at 3.24 h. The significance of the curves are the same as in Fig. 4.

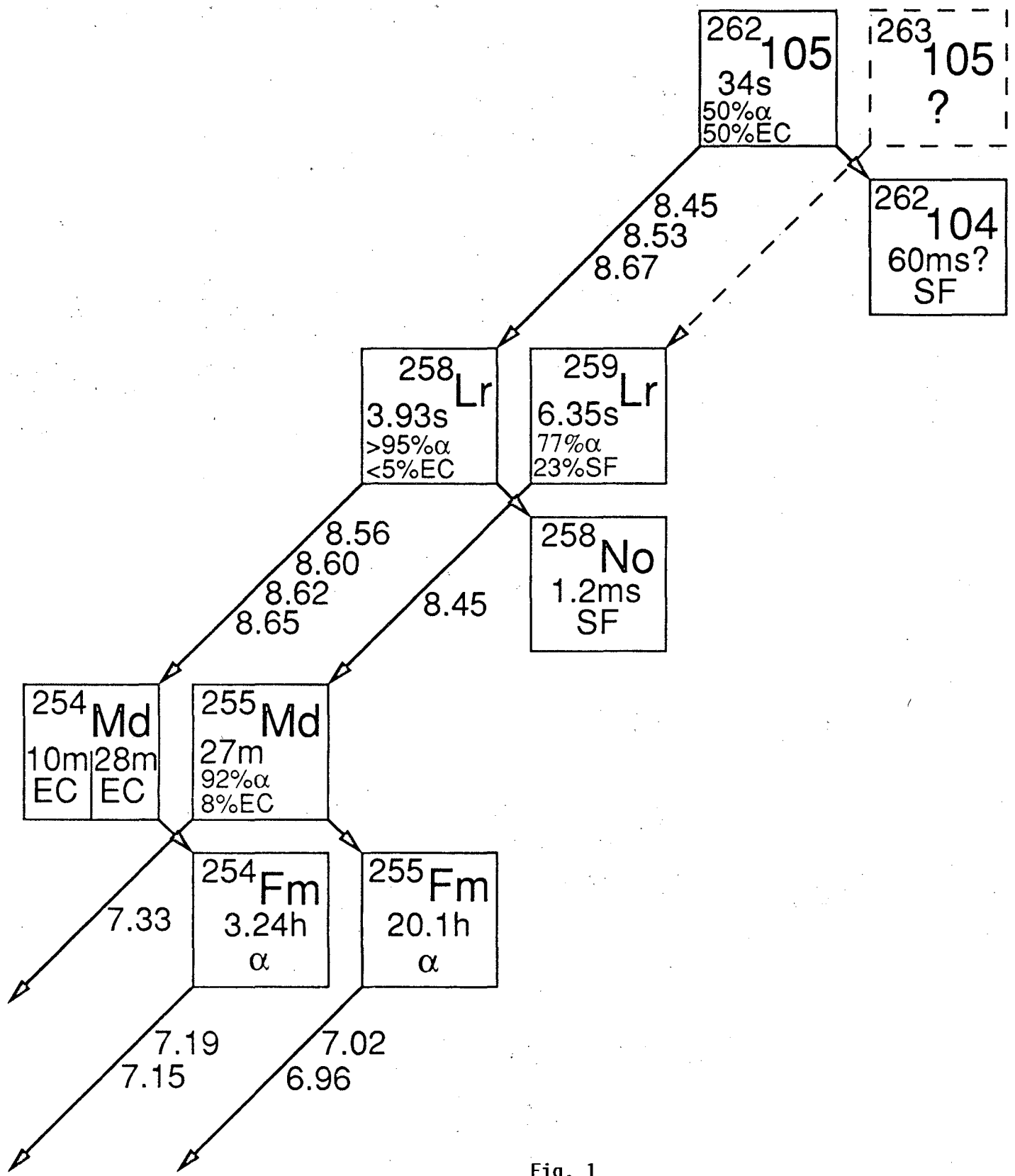
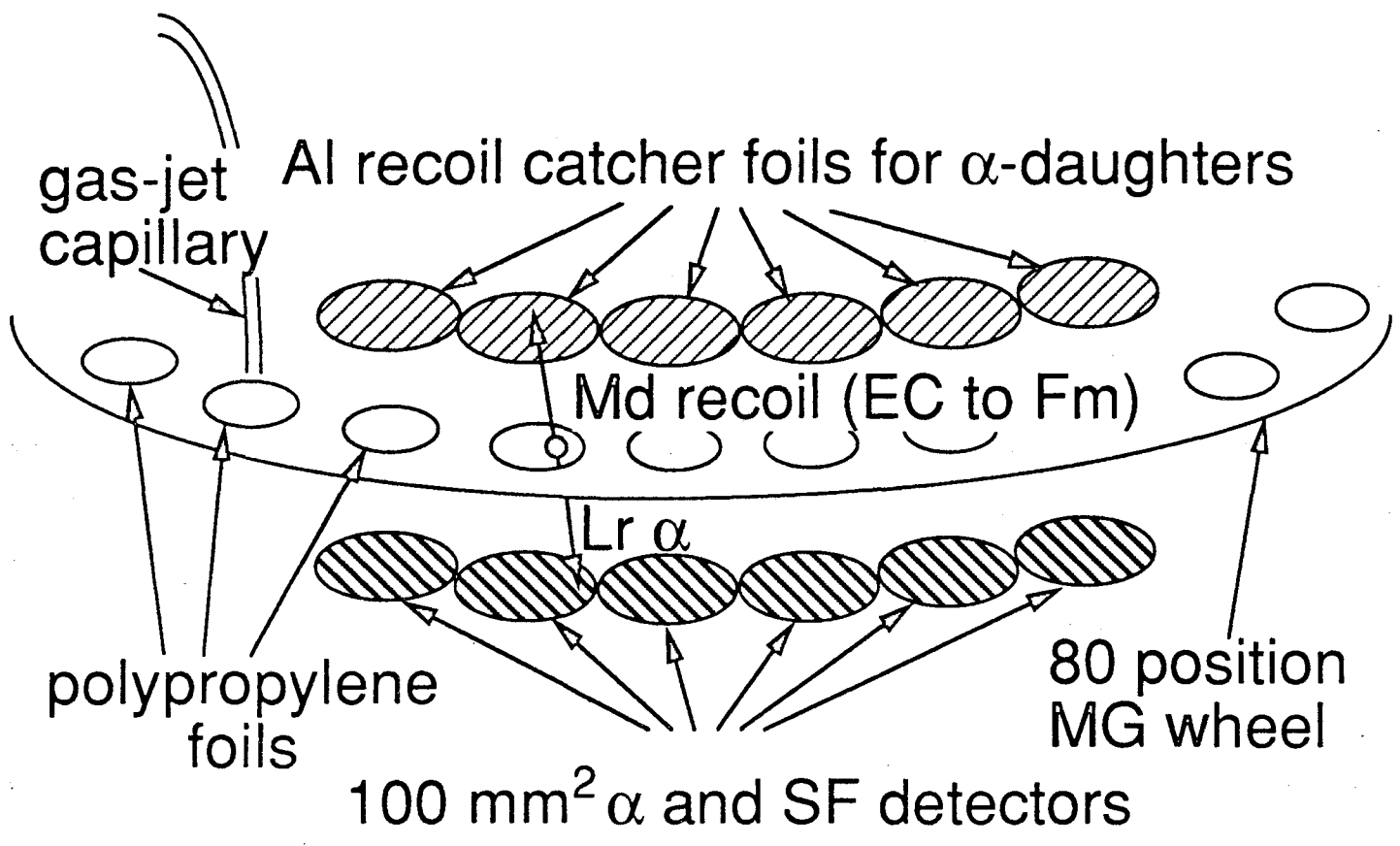
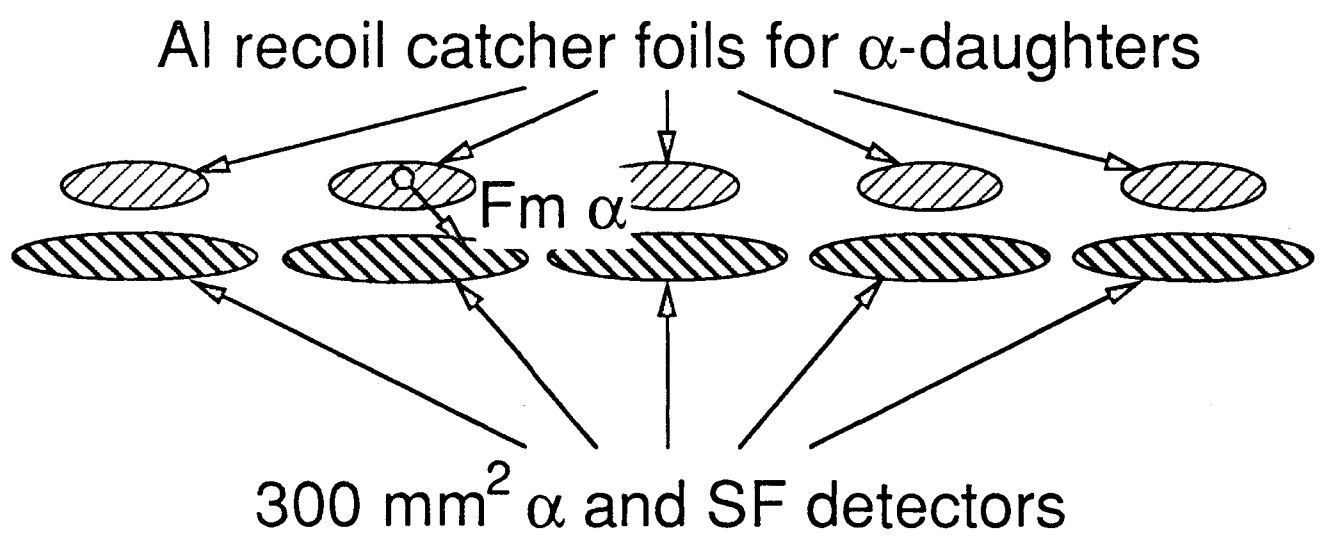


Fig. 1



on-line



off-line

Fig. 2

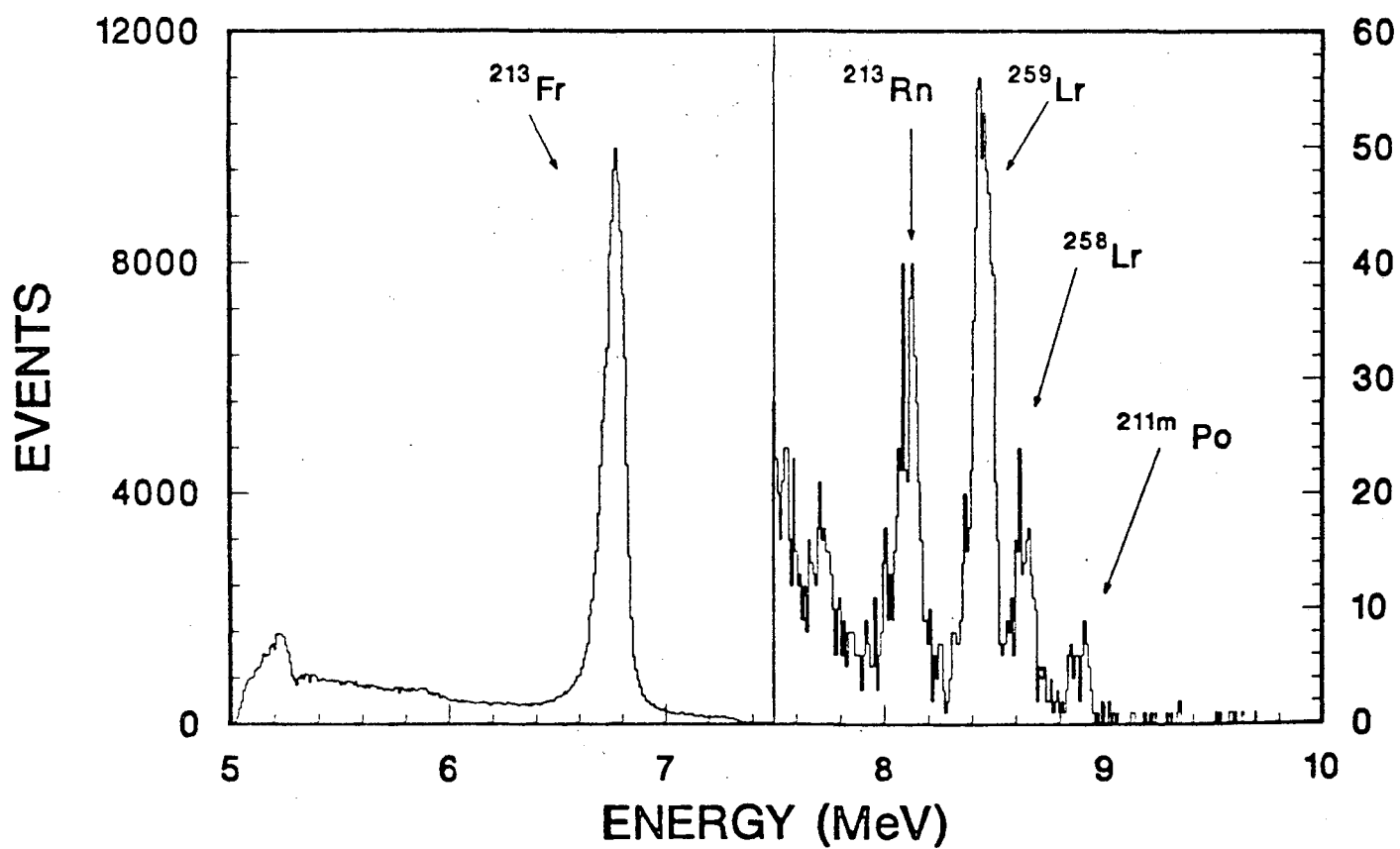


Fig. 3

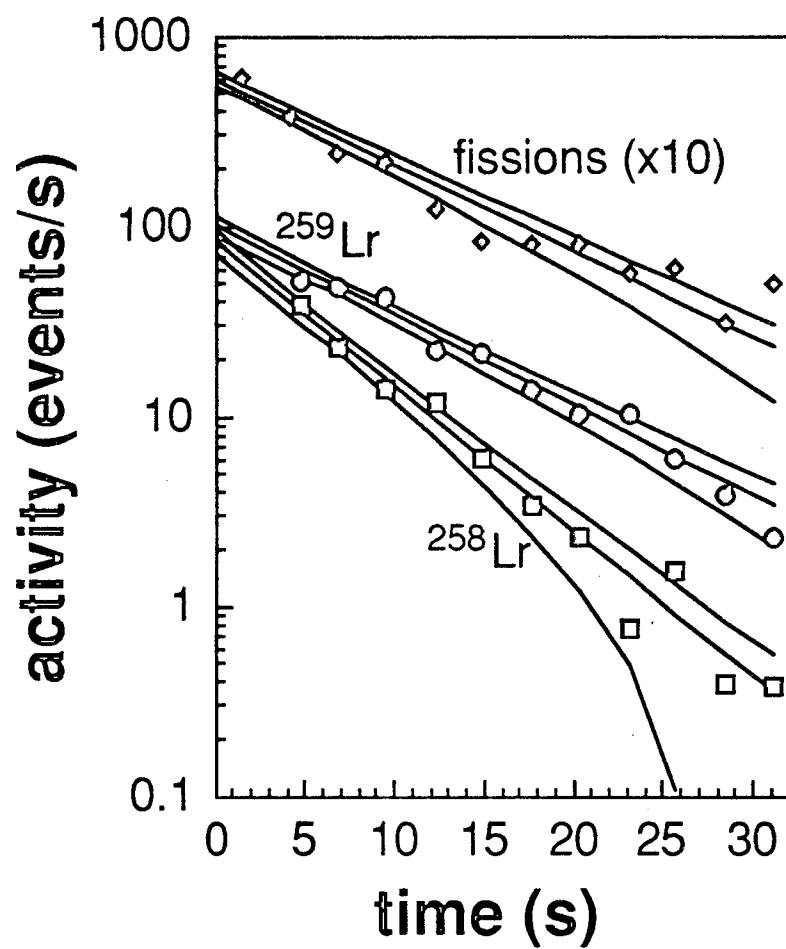


Fig. 4

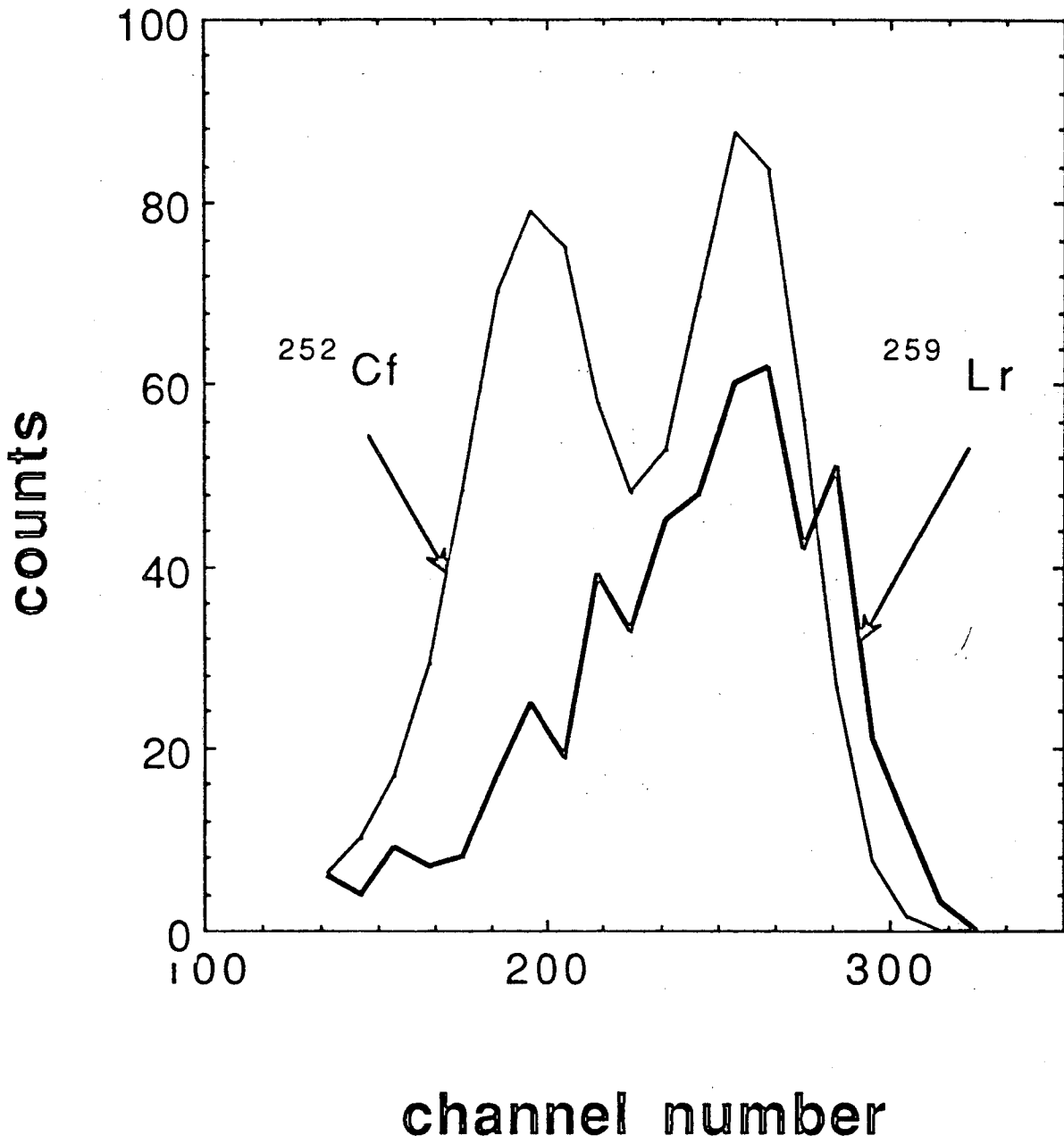


Fig. 5

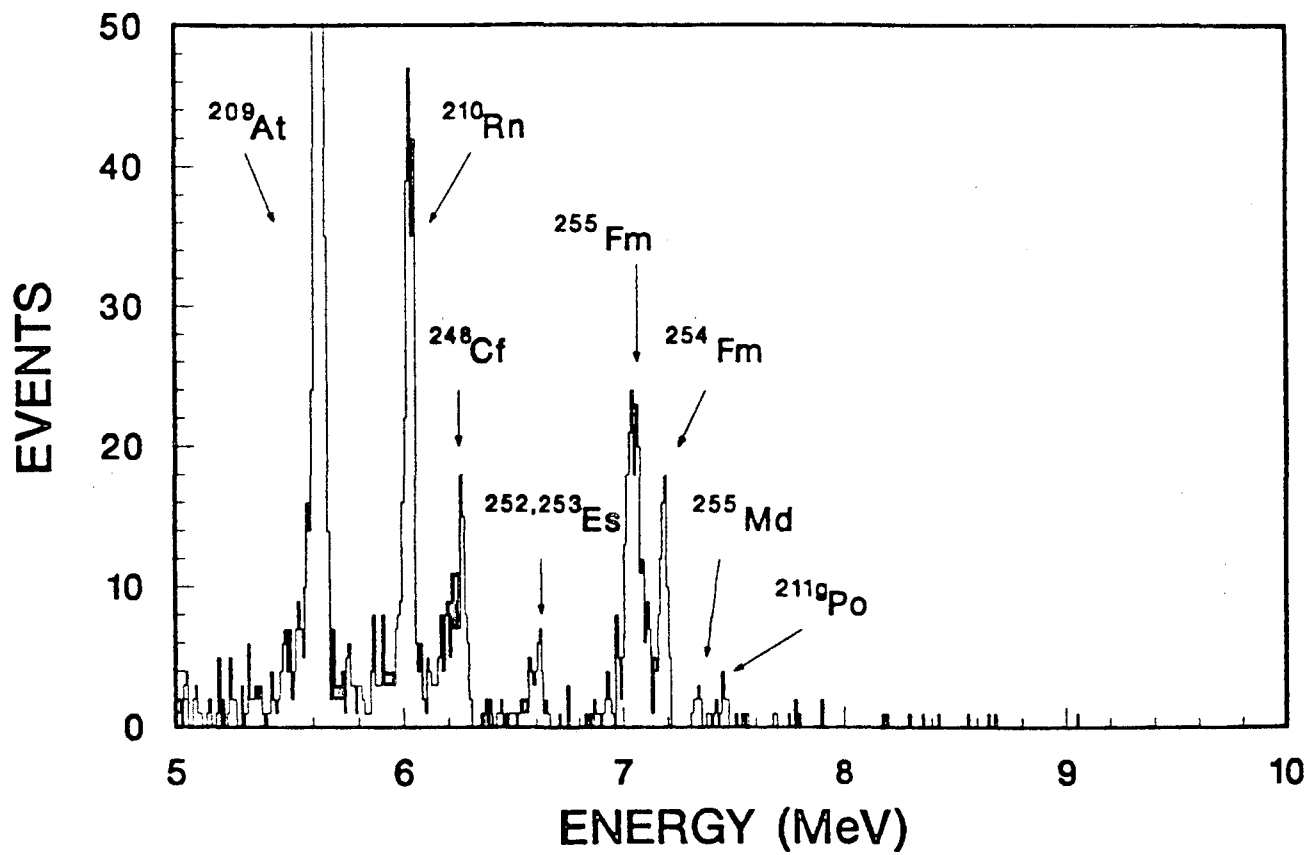


Fig. 6

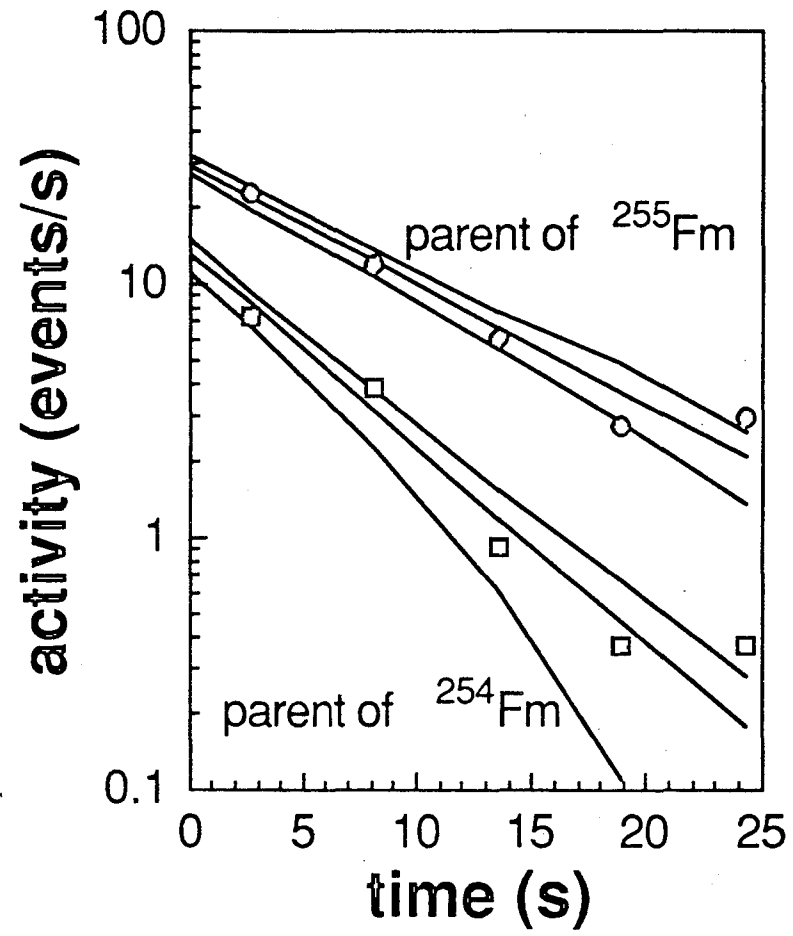


Fig. 7

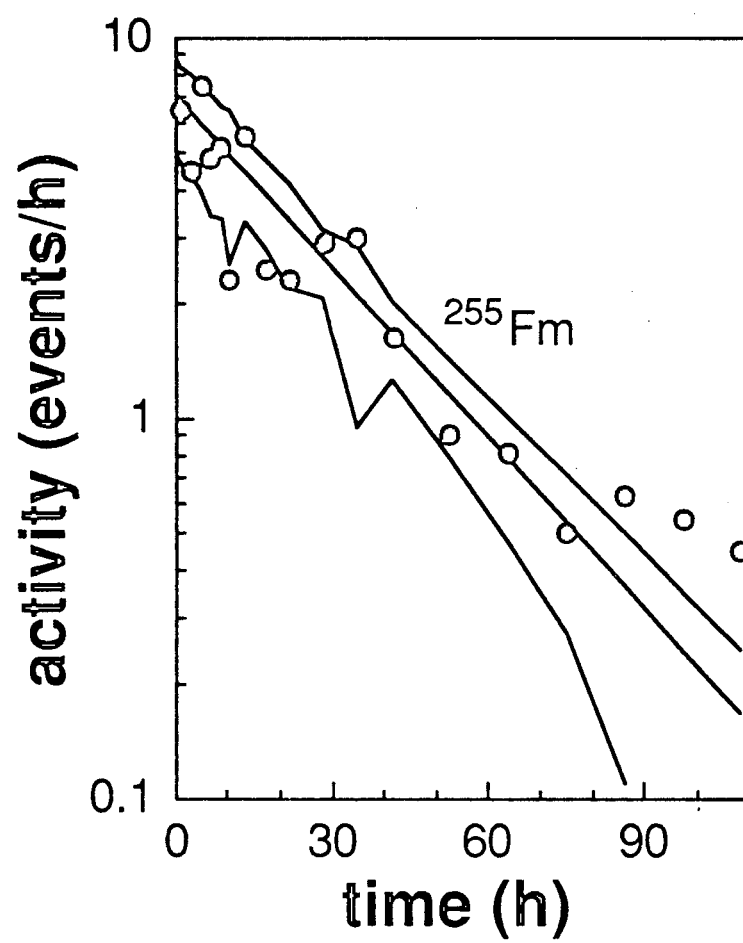


Fig. 8

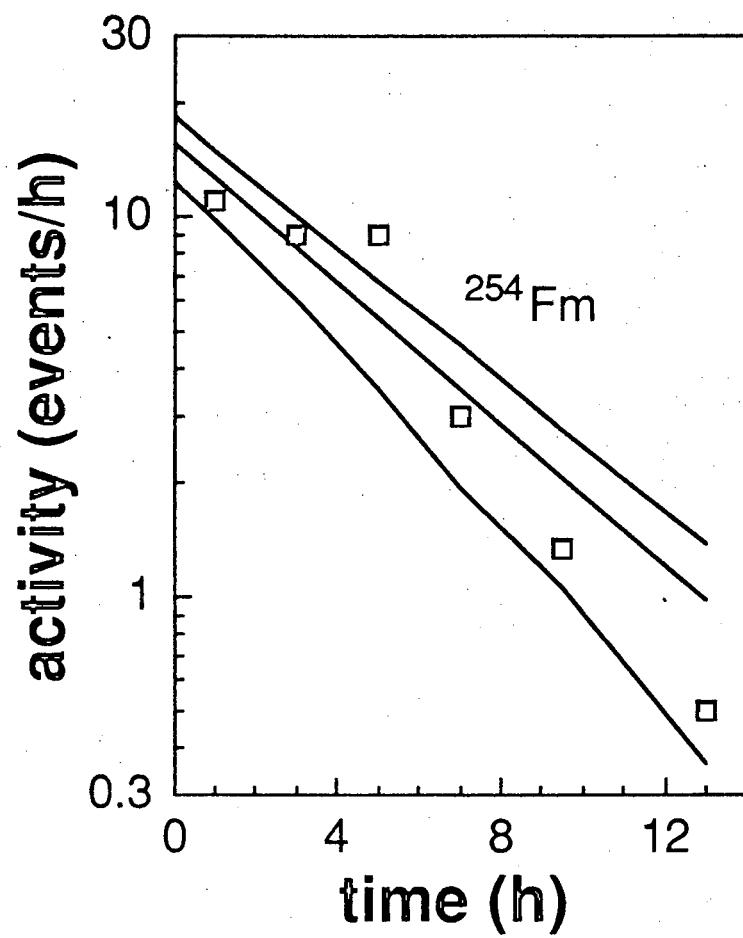


Fig. 9

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