Title

IN DEFENSE OF THE BERKELEY WORK CONCERNING THE ALPHA -EMITTING ISOTOPES OF ELEMENT 104

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September 15, 1970

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UNIVERSITY of CALIFORNIA BERKELEY

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DISCLAIMER

IN DEFENSE OF THE BERKELEY WORK CONCERNING THE ALPHA-EMITTING ISOTOPES OF ELEMENT 104*

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September 15, 1970

INTRODUCTION

Certain questions have been raised regarding the validity of our work reported in Physical Review Letters[1] concerning the discovery of two alpha-emitting isotopes of element 104, rutherfordium. Doubt was first expressed by V. A. Druin in the International Conference on Nuclear Reactions Induced by Heavy Ions[2] at Heidelberg, Germany in July, 1969. These doubts were repeated in a paper submitted to Yadernaya Fizika[3] in November, 1969 by Akapiev et al. In this same month the Robert A. Welch Foundation Conference on Transuranium Elements-the Mendeleev Centennial was held at Houston, Texas and the issues were discussed at length following the presentation of papers[4] by A. Ghiorso and by I. Zvara. Apparently this discussion did not sufficiently clarify the matter because strong doubts were again published in Atomnaya Energiya by G. N. Flerov[5].

In our paper[6] on the discovery of an alpha-emitting isotope of element 105, hahnium, we carefully included answers to the major criticisms, but in view of the continued charges made by the Dubna group we feel that a detailed rebuttal is in order. This paper will attempt to perform this function and in addition will present new evidence in support of our original work.

EXPERIMENTAL METHOD

The research on the heavy-element alpha emitters is fraught with the hazards of low production cross sections which lead to numerous possibilities of misidentifications because of the presence of background activities with similar alpha energies and half-lives. Having pioneered in this field we are painfully aware of these problems and have devoted much of our efforts to determining and eradicating sources of troubles. We feel that we have been conservative in our published claims and are thus much surprised by some of the criticisms leveled at our work. Perhaps we have misjudged the necessity of including a very detailed discussion of possible background effects.

A review of the general methods used in these experiments will help the reader to understand the later discussion of our results. The basic technique takes advantage of the large forward recoil imparted in a heavy-ion reaction to separate the transmuted atoms from the usually highly-radioactive target. In order to provide a very thin sample for alpha particle energy analysis the recoiling atoms are stopped in helium gas at a pressure of 500-1000 torr and then allowed to escape with the gas through a small orifice into a rough vacuum where they impinge and stick upon the rim of a 50 cm diameter wheel. The wheel acts as a conveyor and periodically carries these deposited atoms to positions adjacent to solid-state silicon alpha detectors for energy analysis. Half-life information is obtained from the relative numbers of alpha particles observed at each detecting station and also from the decay observed at each station while the wheel is stationary. The data are stored by a PDP-9 computer and ancillary equipment.

In favorable cases it is possible to determine the mass and atomic number of the nuclide being studied by identifying the daughter nucleus which

recoils from the rim of the wheel as the result of alpha particle decay of its mother. To make this measurement possible the detectors which have been adjacent to the wheel are shuttled about 2 cm to positions opposite similar detectors at suitable intervals so that they can be examined at high geometry for the daughter activity. In our first published experiments on element 104 we used four mother and four corresponding daughter detectors. Later, in order to recover the time lost while the mother detectors were shuttled off the wheel, the detector system was duplicated so that the mother and daughter measurements could be made simultaneously. Thus, at each station four detectors were installed, two shuttled and two unshuttled. The number of detecting stations was first increased to five and finally to seven so that our present system makes use of $7 \times 4 = 28$ detecting crystals as shown in Fig. 1.

CRITICISM AND REBUTTAL

With this general explanation of the method used in our experiments let us take up the criticisms raised in the paper of Akapiev et al[3]. We will first present what we believe to be a fair statement of the major charges raised by that paper and then follow it with our answer. To help the reader follow the arguments we have reproduced two figures (Figs. 2 and 3) along with their captions from the original publication.

1. The Berkeley Group Did Not Present Alpha Spectra Of Background Activities That Would Be Produced By The Bombardment Of Lead Impurities With Carbon Ions And Did Not Discuss Any Background Effects.

These statements are both true. Because of the stringent word and figure limitations set by Physical Review Letters there was no detailed discussion of this aspect of the experiments. Note, however, that we carefully stated that we had made bombardments with other ions and thus implied that suitable

background checks had been made. The fact that certain Pb-produced peaks in the alpha spectra such as ^{214}Ra and $^{209, 210, 212}$Rn were labeled also implies that we were clearly aware of background effects that are caused by a lead impurity. The simple fact is that the element-104 alpha activities were so clearly above the backgrounds usually encountered that this problem was almost irrelevant. Thus we devoted the limited amount of space available to other more important phases of the experiment.

2. Well-Known Pb-Produced Nuclides Do Not Decay With Their Proper Half-Lives, e.g., 30-min ^{209}Rn And 2.7-hour ^{210}Rn.

The main activity in the 6.043 MeV-labeled peak is due to ^{210}Rn and arises as the alpha-recoil daughter of 2.6-sec ^{214}Ra which is produced from a tiny lead impurity in our californium target. Since the Rn isotopes will not stick to the wheel as they emerge from the gas jet (this can only happen if the collecting surface is cooled to a very low temperature), they can only appear in the spectra either by being detected as gaseous atoms in the wheel housing or by being imbedded into the detector surfaces by nuclear recoil. As we will show, the former condition does not prevail. In the latter case they would appear to have the half-life of the mother atoms since the amount deposited into each detector face would depend upon the wheel-cycle period. If the half-life is measured by the relative amounts of activity seen by each detector the mother half-life would determine its value, but if the decay at each detector is monitored, the daughter's half-life would be measured. Thus it was that the 6.043-MeV peak appeared to decay in the successive alpha spectra with a 2.6-second half-life since it was controlled by ^{214}Ra. There must have been some ^{209}Rn (which has the same alpha energy) present in this peak since it is produced as the daughter of 2.75-min ^{213}Ra, but it was a minor component.

3. The Alpha-Particle Peak at 8.87 MeV Can Only Be Due To The 25-sec ^{211m}Po And Thus Cannot Decay With A 3-second Period.

We were very careful to point out that the origin of the 8.87-Mev peak was uncertain and we did not claim that it was due to ^{257}104. From recent results we find that there are several sources that can contribute to this peak in the alpha spectra.

a) One obvious source, of course, is 25-sec ^{211m}Po, as demonstrated in Fig. 5, where a lead target with some 50,000 ngm/cm^2 has been substituted for the ^{249}Cf target which contains about 20 ngm/cm^2 Pb impurity. The polonium activity has a 25-second half-life both as measured from the relative amounts of activity observed at each station and by the decay while the crystals are stationary.

b) At the time when our work[1] was published we were not able to assign the short-lived component in the 8.87 MeV peak to any known nucleus, as was pointed out in the figure caption (Fig. 2). It was not until we started working on element 105 that we accumulated conclusive evidence that ^{257}Lr has a half-life of 0.7 seconds and a most prominent alpha group at 8.87 MeV as opposed to the Dubna group's[7] claim that it had a half-life of 35 seconds and an alpha energy of 8.5-8.6MeV. By resolving the 8.87-MeV peak of the repeat experiment (to be discussed later) into two components we find that about one half of the activity is due to 0.7-sec ^{257}Lr and the other half to 25-sec ^{211m}Po.

c) It is conceivable that 5-sec ^{257}104 can have a weak alpha group at this energy but this can not be resolved easily because of the interfering activities.

4. All Berkeley Half-Lives In The Heavy Element Region Must Be Questioned Because The Apparent Half-Life Of 30-min 7.43-MeV ^{250}Fm Is Obviously Only A Few Seconds.

For some time the strange phenomenon associated with the peak at 7.43 MeV troubled us greatly. We at first quite naturally assumed that the apparent short half-life of about two seconds was a real measure of the half-life of this peak and so thought it might be due to 0.5-sec ^{211}Po, which has this energy, plus some longer-lived background. We finally discovered that the peak persisted after bombardment ended and decayed with a 30-minute half-life at each detector even when the wheel was removed. Further, it was found that the ^{246}Cf daughter of ^{250}Fm was also observed in each crystal and corresponded roughly to the amount of its parent. It was obvious then that the activity was ^{250}Fm which had been transferred to the face of each crystal and the question now became--how were the atoms lodged there? They could not have been transferred as the result of alpha recoil from ^{254}No for two reasons: (1) the number of atoms required was far in excess of that observed and (2) its half-life is well-known to be 55 seconds[8,9].

Over a period of time we have made additional observations and experiments so that now we can offer the following explanation which appears reasonable. We have found that another isotope of fermium, ^{252}Fm, which was made in good yield at the same time as ^{250}Fm, was not transferred. We also found that ^{254}No was transferred to the detectors with a 0.2 second apparent half-life, even though another isotope of nobelium, ^{255}No, was not. We also looked with high sensitivity but without success for the transfer of the prominent activities ^{214}Ra and ^{213}Fr. We decided that the movement of atoms from wheel to detector was not a mechanical or chemical effect but must be due to the recoil afforded by transitions from isomeric states in the two nuclides, ^{250}Fm and ^{254}No. To prove that this was the case we biassed the wheel negative by 10 volts relative to the crystals and increased the gas pressure in this region to 5-10 torr.

By this technique we were able to reduce the transfer of these activities by an order of magnitude.

A very strong corroboration was furnished by the bombardment of ^{242}Pu by ^{12}C ions. At 72 MeV, which is near the peak of the $^{12}C,4n$ reaction, the transfer of 30-min ^{250}Fm was observed with the same apparent half-life as in the carbon ion bombardment of ^{249}Cf. This experiment rules out the rather remote possibility that a 2-second ^{250}Md, decaying by electron-capture, can be the source of the transfer of ^{250}Fm since elements with atomic number greater than 100 cannot be made in this bombardment. It is also extremely unlikely that a 2-second beta-decaying isomer of ^{250}Es can be responsible. We can only conclude that an isomer exists in ^{250}Fm itself and a final test which is to be conducted will be with the reaction $^{249}Cf(He^4, 3n)^{250}Fm$. A full report on this very interesting nuclide will be published later.

With only the evidence afforded by this unusual transfer phenomenon the Dubna group suggested that all of our half-life measurements could be in error because of some undiscovered artifact of our system. Suffice it to say that we have obtained the same values for many nuclides as they have and most of these half-lives were measured before their published work. These measurements vary in range from 3.2 seconds[9] for ^{256}No (which from 1963 to 1967 was quoted as 8 seconds by its Russian discoverers[10]!) to 200 seconds[11] for ^{255}No. Aside from statistical considerations we have found no reason to question our decay periods.

5. The Construction And Geometry Of Our Setup Causes "3 seconds" To Be A Characteristic Value In Our Measurements. In Particular, Activity May Be Collected Directly On The Detector Faces By Gas-Jet Leakage Around The Wheel.

To determine whether a small amount of activity could bypass the wheel

and be collected on the detector faces, we have performed an experiment to measure such an effect. Under our normal conditions some 2 x 10^5 alpha counts of ^{214}Ra were observed from carbon ion bombardment of a lead target with the wheel being cycled at a rate of 3 seconds per detecting station. The wheel was then stopped and the measurement repeated for an identical integrated beam charge. Not one count which could be ascribed to ^{214}Ra was observed in any of the detectors. This was not particularly surprising to us since the volume through which the wheel is rotated is carefully shielded internally by lead sheets to reduce the fast-neutron degradation damage of the silicon detectors. We feel we can state categorically that in our system there is no significant direct transfer by a gas leakage path. Our "3-second half-lives" can only be the result of nuclear characteristics beyond our control.

6. <u>The Half-Lives Of ^{257}104 And ^{259}104 Could Be Much Shorter Because There Is An Excess Of ^{253}No And ^{255}No On The First Detector.</u>

This criticism must be answered in two ways.

a) If there was such an excess of these two isotopes on the first detector, then why would not the alpha particles from their presumed parents be also observed in this same detector?

b) We never said that there was an excess of either ^{253}No or ^{255}No in the first detector. The excess activity was identified as ^{254}No (from its decay on the detector with a 55±20 second half life). We used this isotope of nobelium as a monitor of the amount of "self-transfer" in the daughter-measuring experiment. It now appears that this transfer was accomplished by the decay of the ^{254m}No I.T. with a 0.2-second half-life as explained above.

7. <u>The Berkeley Experiment Can Be Compared To A Background Run Made At Dubna With A Lead Plus Carbon Ion Bombardment And This Shows An Over-Whelming Ratio Of ^{211m}Po To ^{214}Ra.</u>

The Russian experiment is by no means a suitable stand-in for our experiments on element 104. They used a lead target weighing 1500 micrograms/cm^2; we used a californium oxide target weighing about 300 micrograms/cm^2. They used a mixture of helium and nitrogen gases in their recoil chamber; we used only helium. Their recoil chamber dimensions and gas pressure are not given but we suspect that they must be using one much larger than ours at a higher pressure and thus capable of stopping more noncompound-nucleus recoils. It has been our experience that the yield of ^{211m}Po depends critically on these parameters and on other factors such as the impurity content of the carrier gas--even beam level can play an important role. Our experiment has been carefully tailored to maximize the heavy nuclide yield and reduce that of interfering activities so it should not be too surprising if our ratio of ^{211m}Po to ^{214}Ra is lower than in their experiment. We do know that there is some ^{211m}Po in the 8.87-MeV peak but it has not affected the interpretation of the nearby alpha groups which belong to ^{257}104. It is clear that the Dubna system operates differently from ours because of the large amount of ^{213}Rn (8.09-MeV peak) visible in their spectrum. It appears to us that this can only be observed by decay from gaseous atoms of radon in the vicinity of the detectors since the amount is hundreds of times larger than that which could come via electron-capture branching from the 6.77-MeV ^{213}Fr, no trace of which is visible in their spectrum.

NEW EXPERIMENTS

The above statements with their rebuttals we believe to be a fair summary of the criticisms leveled at our experiments. A more satisfying answer, perhaps, is furnished by the following.

We have repeated the ^{249}Cf(^{12}C,4n)^{257}104 $\longrightarrow$ ^{253}No experiment with a

higher efficiency and better resolution. A background experiment was also performed with a 50 microgram/cm^2 lead target. This was not exactly a stand-in for the californium experiment, serving only as a reasonable approximation, but showed that ^{211m}Po was not an important problem. The new element-104 experiment gave statistically better information than the previous run even though it was run in a shorter time.

The results of the repeat experiments are presented in Figs. 4, 5 and 6. The first one displays a series of alpha-particle spectra of the activities produced in bombardments of the ^{249}Cf target with 74-MeV ^{12}C ions. The experiment was done with the 20-detector system and the spectra shown are from the detectors in the on-wheel position. The wheel-cycle rate was 3 seconds per detecting station. The activities assigned to isotopes of Ra, Fr, Rn and Po are due to a lead impurity in the target. A small mercury contaminant in the target-backing material does not significantly add to these activities. In cases where two activities contribute to the same peak the approximate ratio of the two is shown below the peak label. It should be noted that the 6.26-MeV peak is predominantly due to ^{248}Cf, not due to ^{212}Rn as indicated in Fig. 2. The 6.04-MeV ^{210}Rn peak has the apparent half-life of its mother, ^{214}Ra, because the observed events are mostly due to the decay of ^{210}Rn atoms recoiled into the detectors as a result of the alpha decay of ^{214}Ra atoms.

The peak at 8.87 MeV results from the decay of ^{257}Lr and ^{211m}Po, both of which contribute about equal amounts. The assignment of the 0.7-sec component to ^{257}Lr was confirmed in connection with the experiments on element 105 by a cross-bombardment utilizing the ^{249}Cf(B^{11},3n)^{257}Lr reaction.

Fig. 5 displays a series of alpha-particle spectra of the activities

produced in bombardment of a natural lead target by ^{12}C ions under similar conditions to those in Fig. 4. The ratio of $^{210}Rn/^{214}Ra$ is smaller than in Fig. 4 because the decay of ^{210}Rn was not followed equally long after the bombardment. Because of the differences in target thicknesses and beam intensities, and about 1.5-MeV difference in the bombarding energy of the ^{12}C ions, the ratio of $^{211m}Po(8.87\ MeV)/^{214}Ra(7.14\ MeV)$ is higher by a factor of two in the lead spectrum.

The series of alpha-spectra from the same experiment as in Fig. 4, but recorded by the crystals in the off-wheel position, is shown in Fig. 6. The on-wheel-off-wheel cycle, or shuttle period, was 100 seconds, while the wheel-cycle rate was 3 seconds. The presence of ^{250}Fm and ^{254}No alpha-particle peaks with apparent half-lives of 2 sec and 0.2 sec, respectively, is believed to be caused by isomeric states of these isotopes, as previously explained. The other peaks in the spectrum are caused by the decay of alpha-recoil atoms on the movable detectors.

The apparent half-life of ^{253}No obtained by measuring the amount of ^{253}No recoiling at each detector station is 4.4 ± 1.0 seconds. This is in agreement with the value 4.8 ± 0.5 seconds for $^{257}104$ as measured directly by the amount of activity observed at each station. Summing the detected daughter events by the four 25-second time subgroups of the 100-second shuttle period, one obtains a half-life of 90 ± 30 seconds for the 8.01 MeV alpha-particle group which is consistent with the 105-second half-life[9] of ^{253}No. The ratio of counts in the alpha-particle peaks assigned to $^{257}104$ to those in the 8.01-MeV peak in Fig. 6 is 506:137=3.5. A calculation taking into account geometry and timing conditions gives a value 2.5. This difference could possibly be an indication of some 20-30% electron capture branching in

the decay of ^{253}No.

The same equipment was used to discover another isotope of rutherfordium. This work[12] characterized the 65-second alpha emitter, ^{261}Rf, and showed conclusively that its 26-second daughter, ^{257}No, was observed as the result of alpha particle recoil-transfer from its mother. This discovery was confirmed chemically when it was shown[13] by fast aqueous-chemistry experiments utilizing this isotope that rutherfordium was a trans-actinide element. More recently this system was used in the discovery[6] of an alpha-emitting isotope of element 105, the 1.6-sec ^{260}Ha. The fact that the radiations detected in these two sets of experiments were completely different shows that there is no built-in constant background effect that could conceivably mislead us in our interpretations of other results.

SUMMARY

We feel that we can safely draw the following conclusions:

1) The additional experiments with ^{257}Rf which have been described above fully confirm our original findings.

2) The puzzling transfer phenomenon has been explained as the result of previously unknown isomeric transitions in the even-even nuclides, ^{250}Fm and ^{254}No.

3) The unknown 0.7-sec 8.87-MeV activity has been determined to be due to ^{257}Lr.

4) There is no gas leakage effect in our system that can transfer activity directly to the detector faces.

5) Our system does not have a "characteristic half-life due to some unexplained artifact".

In our opinion the performance and reliability of our experimental system is beyond reproach. The present multi-detector shuttle apparatus is quite a complicated instrument which has required a great deal of development and testing and its value as a research tool has been proven by the quality of the nuclear spectroscopic data obtained for the seven isotopes of nobelium, five of lawrencium, three of rutherfordium, and one of hahnium which have been characterized up to the present time.

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†On leave of absence from Department of Physics, University of Helsinki, Finland.

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FIGURE CAPTIONS

1. A schematic representation of the vertical wheel system with seven detecting stations. On the right hand side a cross section of one of the detector stations is shown.

2. "A series of alpha spectra of the activities produced by bombardment of ^{249}Cf with 71-MeV ^{12}C ions. The top spectrum is the sum of the individual spectra from the four detectors. The 8.60-MeV peak is probably due to ^{258}Lr; the peaks above that energy belong to ^{257}104 with the exception of the one at 8.87 MeV whose origin is uncertain[1]." (See text of this paper: This peak is now known to be due to ^{257}Lr and ^{211m}Po.)

3. "A set of spectra from the mother-daughter experiment which demonstrates the genetic relationship between ^{257}104 and ^{253}No. The spectra recorded by the individual crystal pairs are shown on top with the sum of the last three pairs at the bottom[1]."

4. A series of alpha-particle spectra of the activities produced in repeat experiments when bombarding ^{249}Cf target with 74-MeV ^{12}C ions. A 20-detector system was used instead of the 8-detector one employed in the experiments resulting in the spectra shown in Fig. 2. The wheel-cycle rate was 3 seconds in both series of experiments.

5. Alpha-particle spectra resulting from an experiment where the ^{249}Cf target was replaced by a lead target.

6. A series of alpha-particle spectra from the same experiments as in Fig. 4 but recorded by the crystals in off-wheel position. These spectra correspond to those displayed in Fig. 3, but show a wider range of alpha-particle energies.

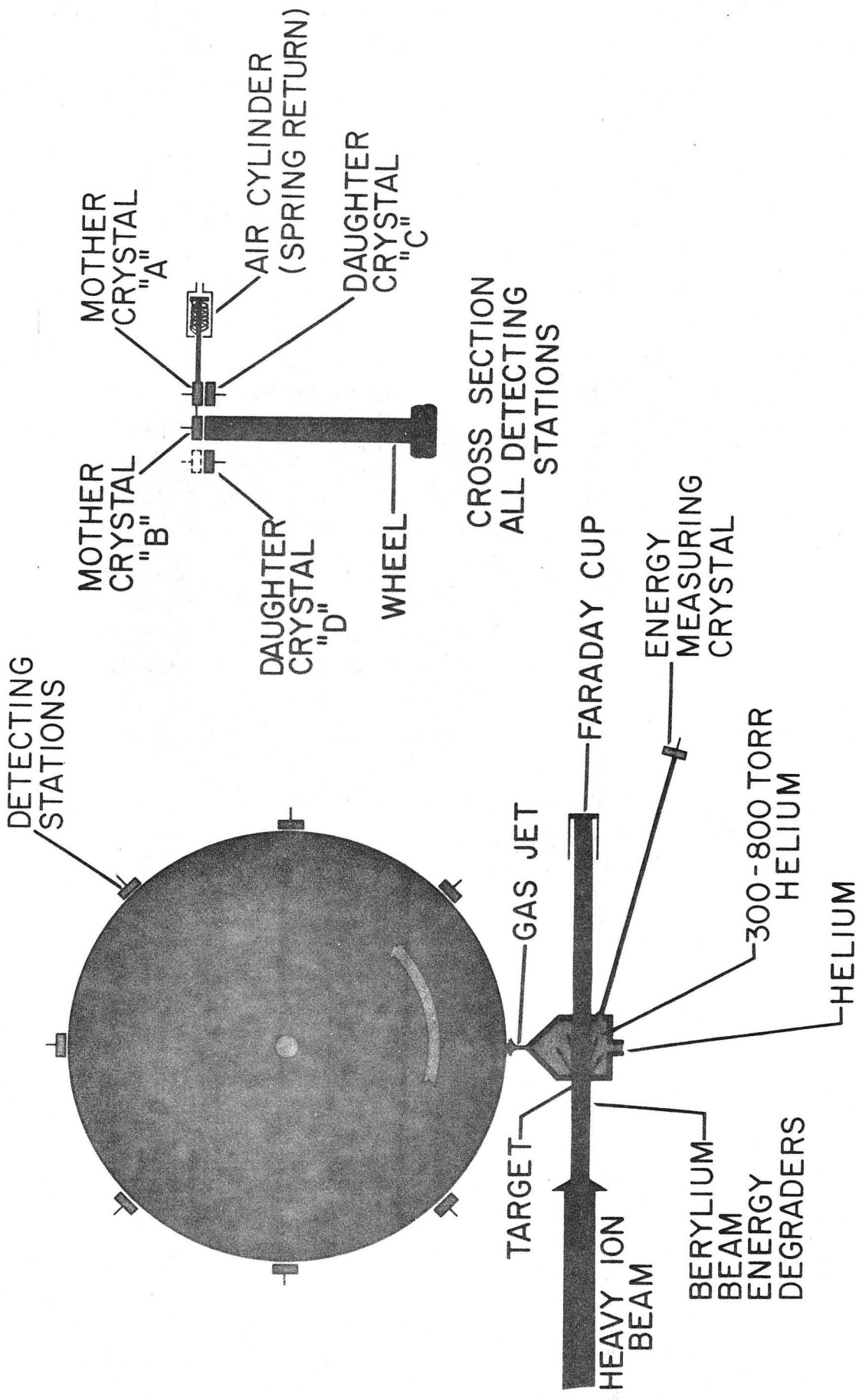
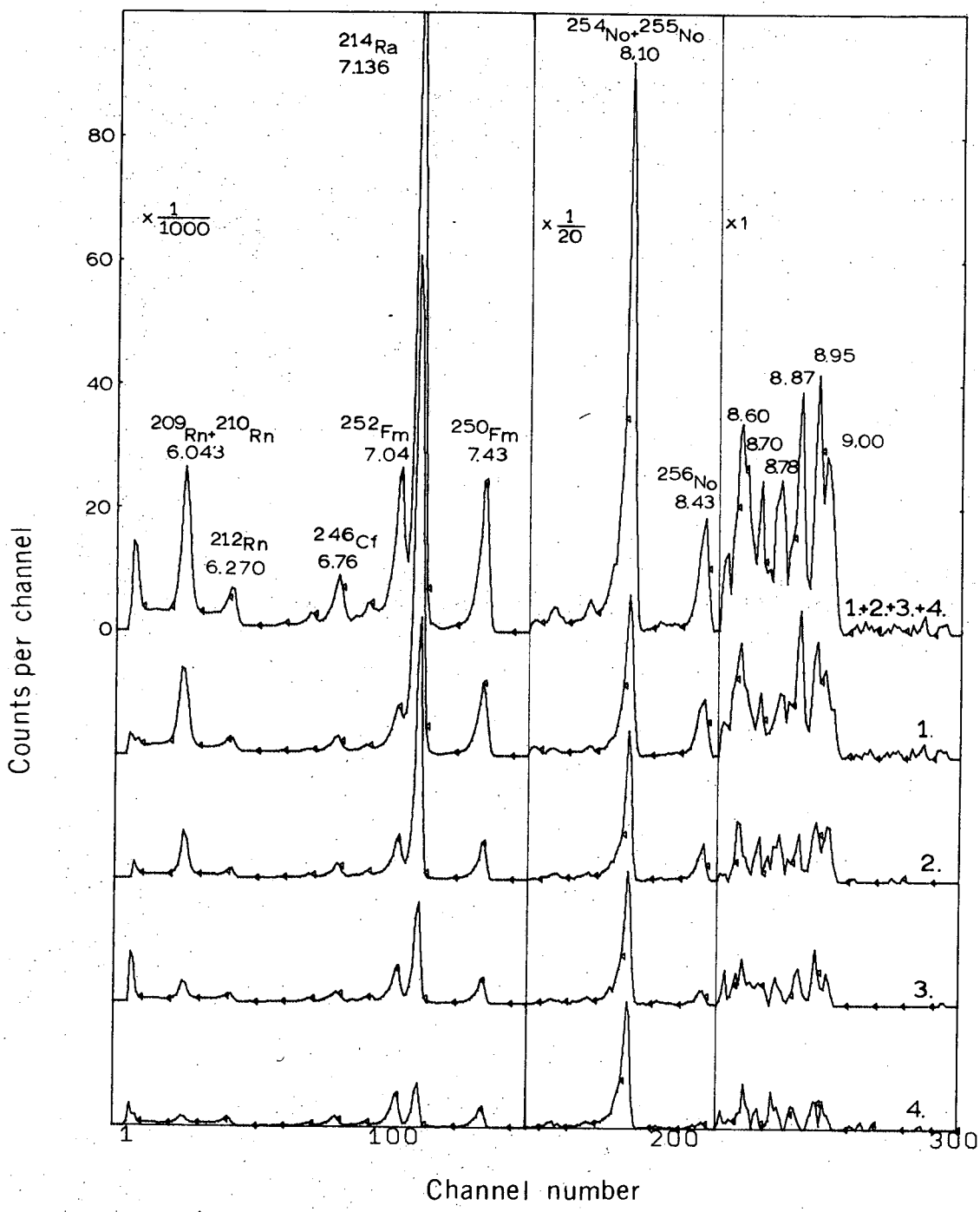

DETECTING STATIONS
MOTHER CRYSTAL "A"
AIR CYLINDER (SPRING RETURN)
DAUGHTER CRYSTAL "C"
MOTHER CRYSTAL "B"
DAUGHTER CRYSTAL "D"
WHEEL
CROSS SECTION ALL DETECTING STATIONS
GAS JET
FARADAY CUP
ENERGY MEASURING CRYSTAL
300-800 TORR HELIUM
HELIUM
TARGET
BERYLIUM BEAM ENERGY DEGRADERS
HEAVY ION BEAM

FIG. 1



XBL 694 4816

Fig. 2

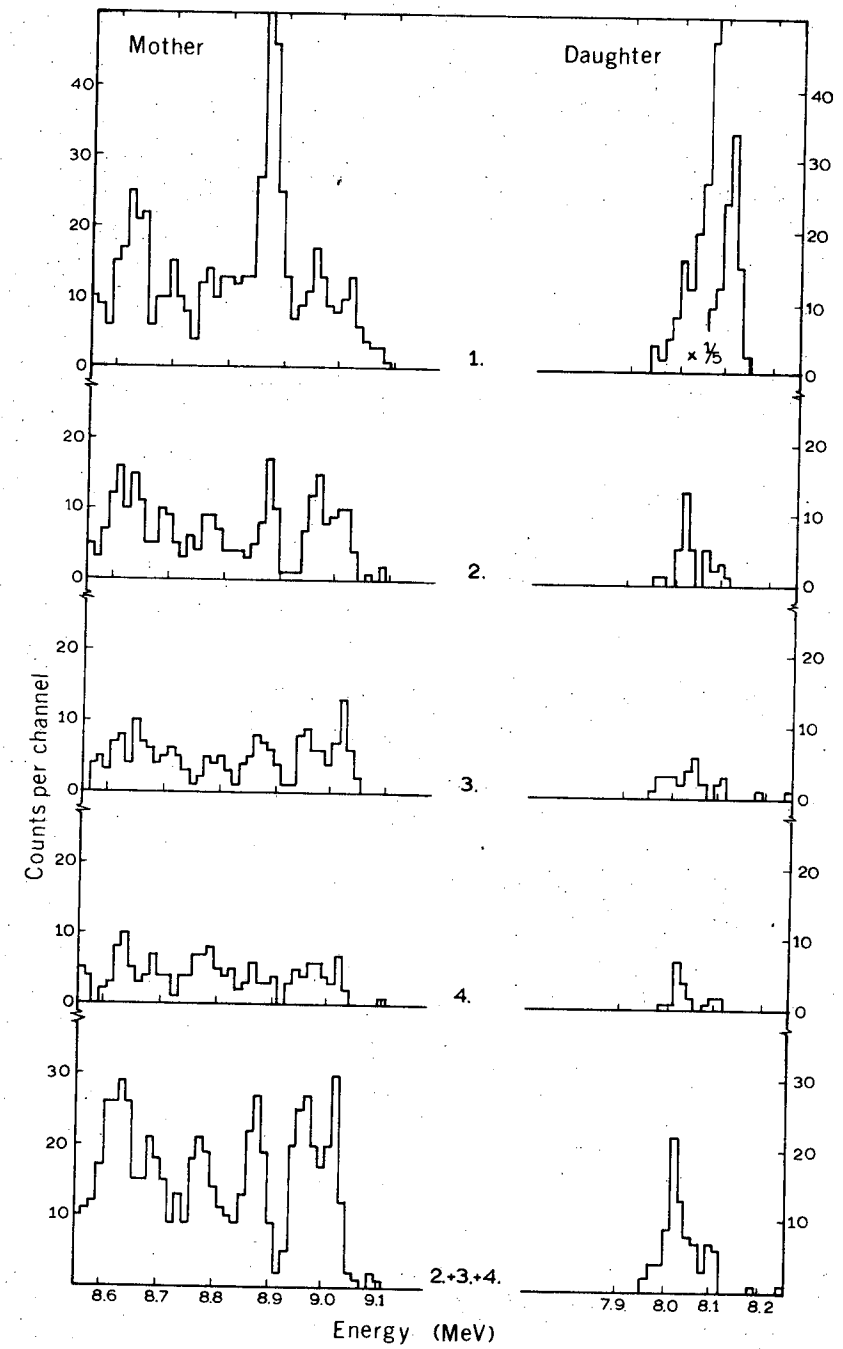

Mother
Daughter
1.
2.
3.
4.
2.+3.+4.
× 1/5
Counts per channel
Energy (MeV)
8.6
8.7
8.8
8.9
9.0
9.1
7.9
8.0
8.1
8.2
XBL 694 4813

Fig. 3

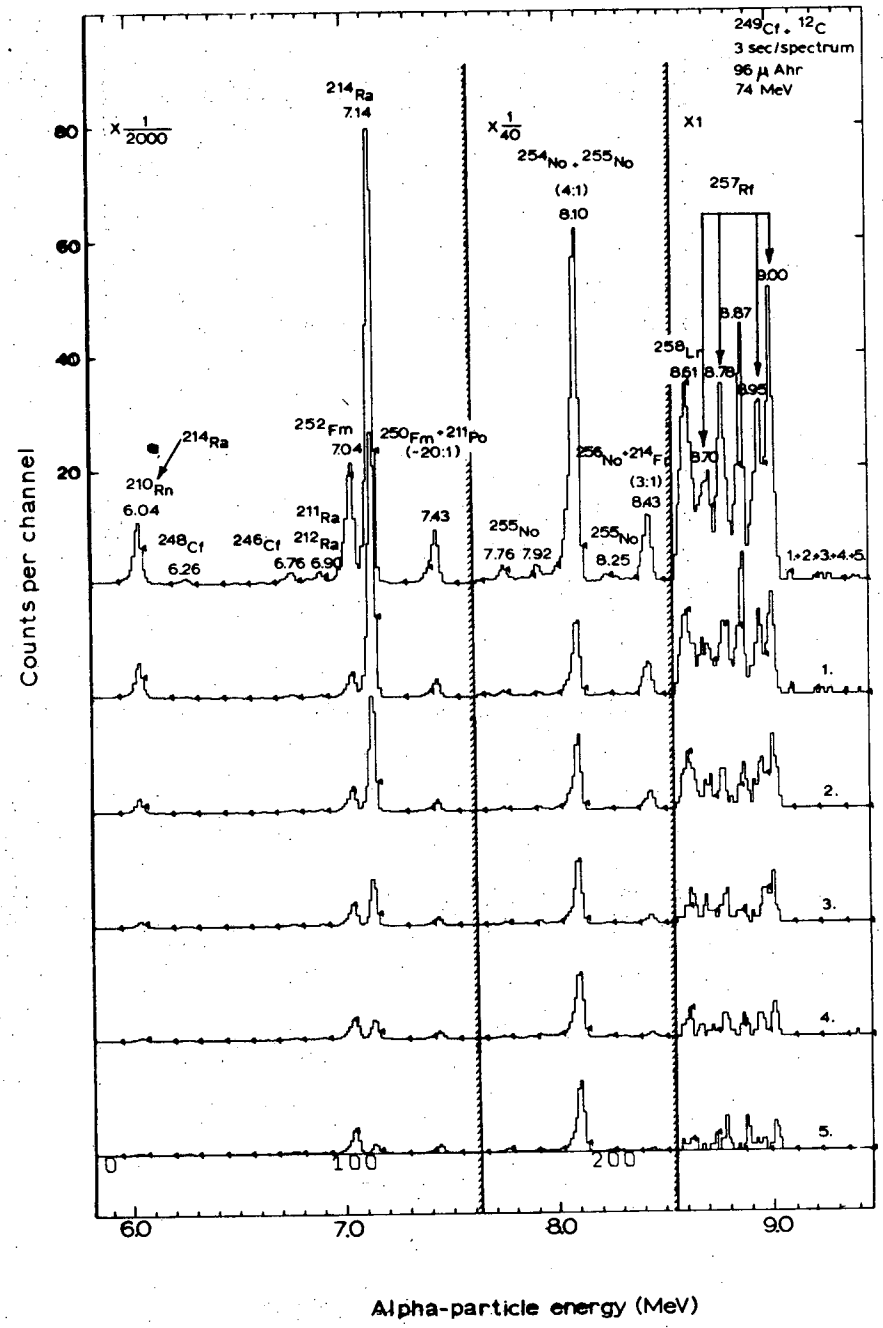


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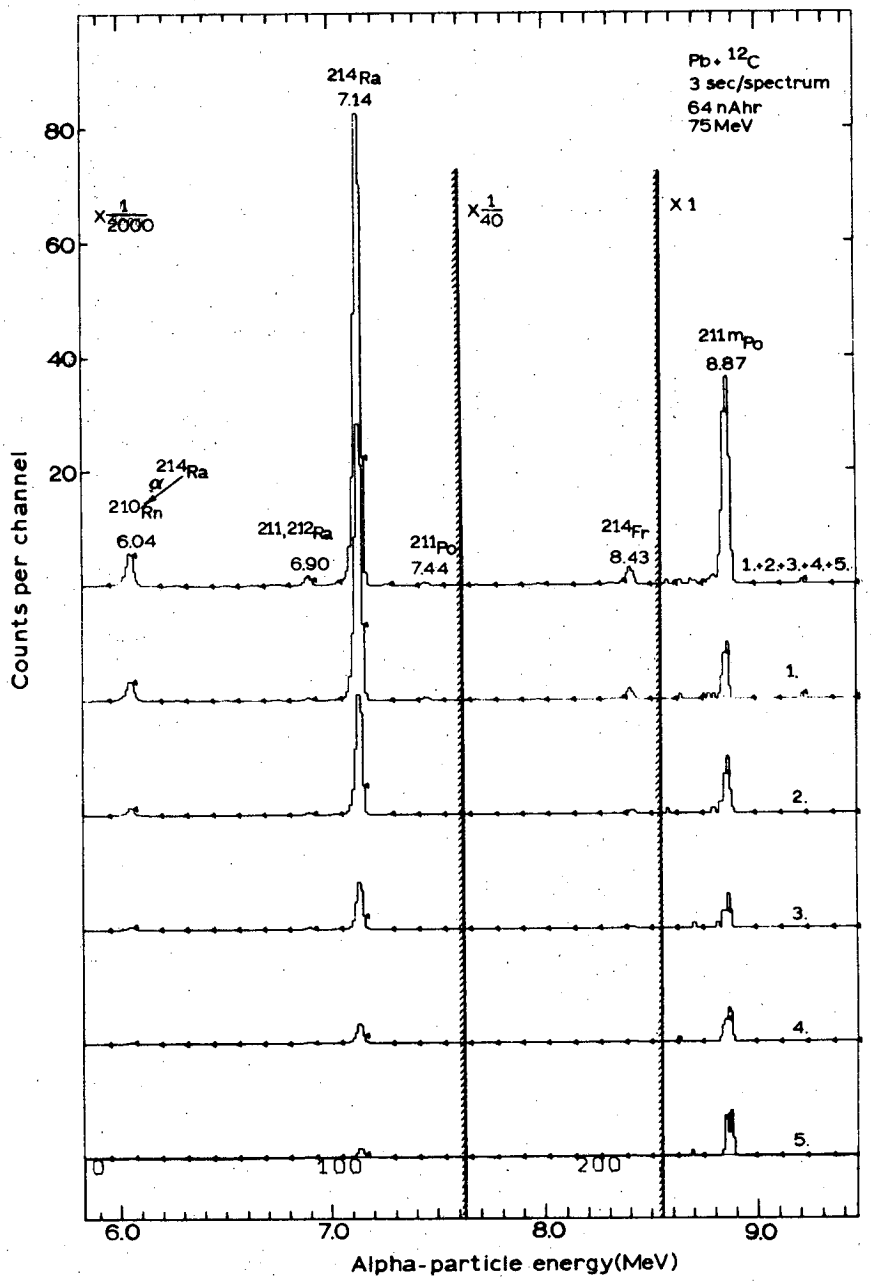
Fig. 4



XBL 702 6139

Fig. 5

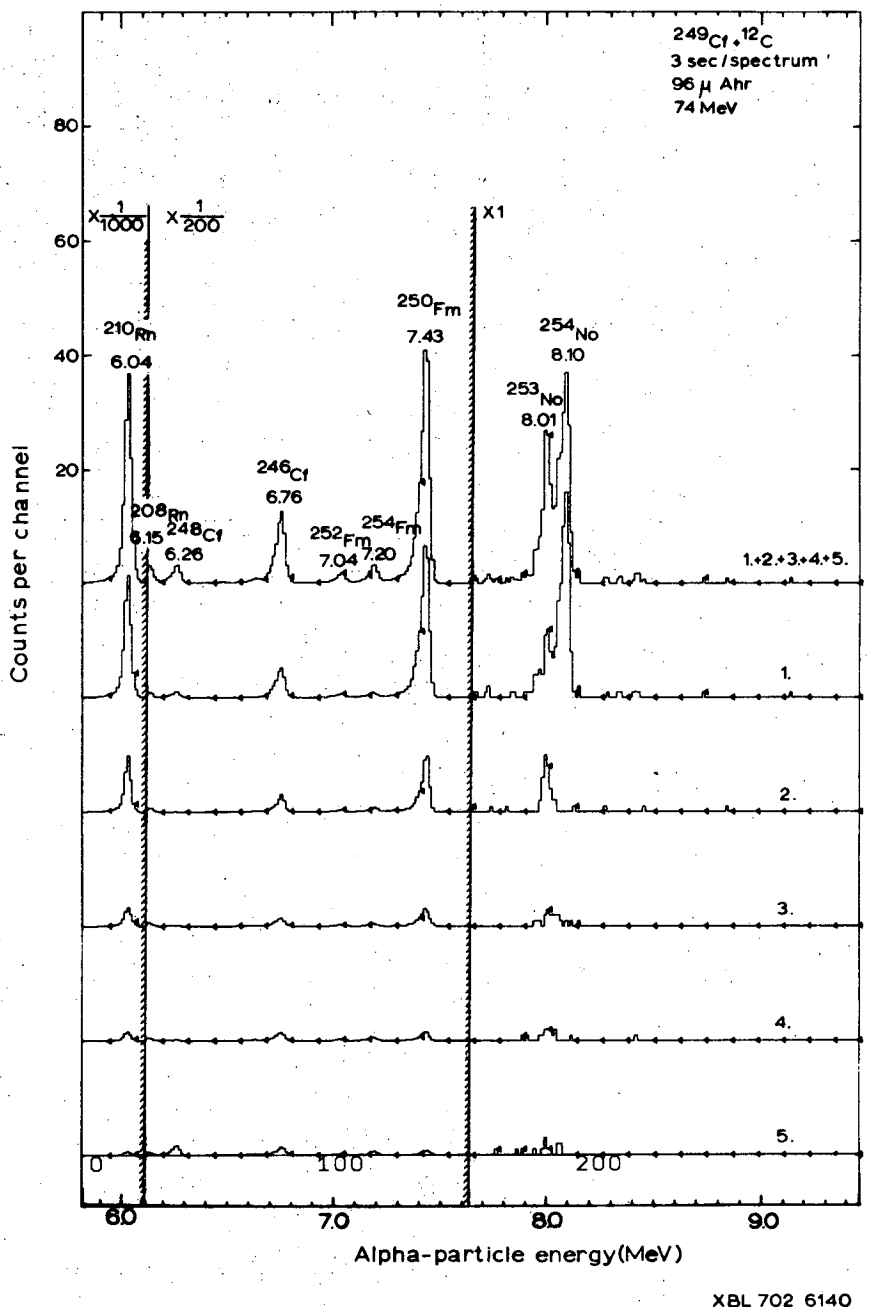

249Cf + 12C
3 sec/spectrum
96 μ Ahr
74 MeV
X 1/1000
X 1/200
X1
210Rm
6.04
208Rm
6.15
248Cf
6.26
246Cf
6.76
252Fm
7.04
254Fm
7.20
250Fm
7.43
253No
8.01
254No
8.10
1.+2.+3.+4.+5.
1.
2.
3.
4.
5.
Counts per channel
Alpha-particle energy(MeV)
XBL 702 6140

Fig. 6

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