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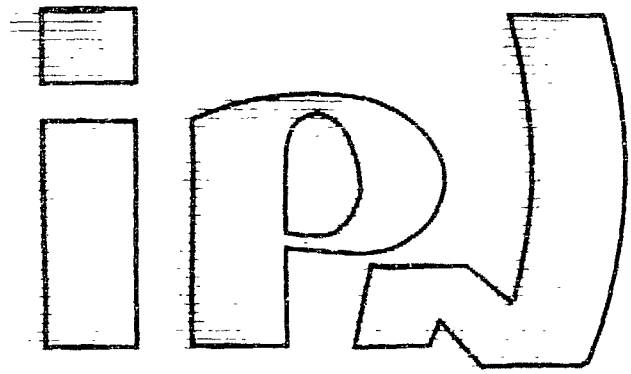

IPNO DRE 90-20

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[×] Institut de Physique Nucléaire, Orsay, France

[+] Institut Laue Langevin., Grenoble, France

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M. Bernas[X], P. Armbruster[+], S. Czajkowski[X], H. Faust[+] and J.P. Bocquet[O]

[X] Institut de Physique Nucléaire, Orsay, France
[+] Institut Laue-Langevin, Grenoble, France
[O] Institut des Sciences Nucléaires, Grenoble, France

Summary

Fragments of mass A=68 and 69 are identified for the first time in thermal fission .Their chain production yields and isotopic percentages are evaluated on ^{239}Pu. The ß-decay half-lives of the very neutron-rich isotopes ^{69}Co, ^{68}Co and ^{68}Fe are measured from time correlations between the selected isotopes and the ß particles of their decay.

P.A.C.S. Number: 27-50+e and 25-85 Ec

Thermal fission (n,^{239}Pu) ; Measured production yields of masses A=68,69 and 73 and half-lives of ^{69}Co,^{68}Co and ^{69}Co.

Introduction

Thermal fission has been shown to provide a way of observing very neutron rich isotopes of Cu and Ni (1). This cold process is indeed appropriate to generate isotopes with a large neutron excess. Those loosely bound fragments are born with a small excitation which restricts the neutron emission. Further, their low kinetic energy prevents them from breaking up by secondary reactions.

On the very asymmetric branch of thermal neutron fission, isotopes of mass between 80 and 70 have been identified and the associated yields, which range between 10^{-3} and 10^{-8}, systematically measured (2).

We have developed a method to measure the ß decay half-life of the new isotopes ^{71}Ni to ^{74}Ni. At the end of their trajectories they are implanted in a solid state detector where they undergo ß decay. The associated half-life is deduced from the time correlations between signals from the implantation of the identified nuclei and their subsequent ß-decays (3).

A study of very asymmetric fission was recently performed on a ^{239}Pu target (4). The production yields were found to be larger larger than on a ^{235}U target and to favour slightly more neutron rich isotopes with less odd-even effect. We have extended these measurements to the very light masses A=69 and 68, for which we report here a first determination of the production yields. These yields are extremely low. Nevertheless, with experimental conditions selected in order to optimize either the absolute fragment counting rate or their rate relative to contaminants, the ß-decay half-life of ^{69}Co, ^{68}Co and ^{68}Fe have been evaluated.

Experiment

The fragments from thermal fission of ^{239}Pu induced by the neutrons from the high flux reactor (ILL Grenoble) are separated by the recoil spectrometer Lohengrin (5). They are focused along discrete A/q focal lines (q being the atomic charge state) as a function of E_{Loh}/q. The ionization chamber (6) gives the measure of the kinetic energy E_{Loh}, from which the q value (and therefore A) is deduced. In the first stage of the chamber the measurement of the ΔE of the fragments provides the Z identification (fig. 1).

On the very end of their trajectories the fragments are implanted in one of the 8 planar Si detectors of 10x10x0.5 mm where the ß energy loss signals ($\Delta E\beta$)

are also measured. Since fragments are implanted only on the surface of the Si detector, the ß detection efficiency, ε, is 0.5 at maximum.

Time correlations between identified fragments and either the first β signal or the few subsequent signals counted in a preselected period t_c are both analysed. Two methods, least square fits of time spectra and maximum likelyhood procedures (M.L.H.) are used. Refined analysis is required here since the number of relevant fragments is low and the background counting rate, (per detector and per second) large, due to the large yields of contaminant fragments which ß-decay in the ionization chamber. The method relies on the stability of the counting rates , which are controlled continuously. The experimental details and methods for analysing time correlations have been extensively described (3).

Yields

The chain and elemental yields have been determined by referring to the A=82 chain yield, which is known from previous measurements at Lohengrin (4). With the same experimental conditions - diaphragm, target thickness etc. - we have compared the counting rates for the line A=68 and A=82 at an energy E_{Loh} = 90 MeV and an ionic charge state of q=17, at the maximum of the charge distribution for A=68. But this state is no longer at the maximum of the distribution for A=82, as it was shown in our study on the ^{235}U target (7). Therefore, a correction factor, taken from this latter study, has been applied to evaluate and compare both counting rates for the most abundant charge state (q=18 for A=82). Those partial counting have been assumed to be proportionnal to the chain yields integrated over q and E . This assumption is reasonable given the large counting uncertainties.

The relative isotopic yields have been deduced from the analysis of the spectra of energy loss in the ionization chamber (fig. 1).

The A=69 and A=68 low rate measurements, both at a same energy and charge states (q=17) could be directly compared to obtain the mass yield ratio. Since for Z-q < 10 the atomic shell closure inhibits the production of an ion [7], the q=17 charge state, selected for the ^{69}Co long duration measurements, gives an absolute counting rate for Co isotopes twice as large as that with q =18 (Table I). For this ionic state both mass lines A=69 (q=17) and A=73 (q=18) are simultaneously selected by Lohengrin as they occur with very close values of A/q. Therefore the A=73 chain and isotopic yields have been also measured With a counting rate larger by an order of magnitude, this line provides a convenient monitor of the experiment.

The few results on mass and isotopic yields appear in table 1. They confirm that the mass distribution is wider with a ^{239}Pu than with a ^{235}U target, the slope of the decreasing yields being less steep.The Z distribution for A=73 is also found to be wider with ^{239}Pu and shows less odd-even effect.(see ref.2) Therefore, the special features reported for Cu and Ni isotopes produced in thermal fission of ^{235}U, i.e. large proton and neutron odd-even effect on the yields and the 2 MeV fall in the kinetic energy of these light fragments are indeed to attribute to this fissioning nucleus.

β-Decay Half-life of ^{69}Co, ^{68}Co and ^{68}Fe

The construction of time correlations requires a good identification of the parent fragment - this condition is fullfilled (fig. 1)- and an accurate knowledge of the correlation effectiveness, ε,and of the background rate of the β detector, b. The parameter ε should not take only the β-detection efficiency into account but also the electronics and data-processing. Evaluated systematically on the β-decay of ^{96}Sr ($T_{1/2}$ = 1s) (3) the value of ε has been found to be lower than in previous measurements (ε ≅ 0.4).

In spite of this loss of efficiency and of a larger rate of the β background (b ≅ 0.35 s^{-1} instead of 0.2 s^{-1}) the half-lives under study were still measurable

since they are predicted to be shorter than in our previous work (3) 0.2 s instead of 1 s. But the half-lives of the Ni isotopes, which are mainly produced in the 68, 69 and 70 chains, are out of reach. A reduction of the background rate by more than an order of magnitude would be required to measure these longer half-lives.

a ^{69}Co

Two series of measurements were performed to evaluate the ^{69}Co half-life (table II). They were analyzed separately since the experimental conditions (consequently ε and b) had significantly changed. Both analyses using the two methods lead to compatible results ; $T_{1/2}$ = 0.27 ± 0.05 s. The results are summarized in table III and an illustration is given in fig. 2.

b mass 68

An example of the isotopic distribution at mass A=68, reported on Fig. 1, shows besides ^{68}Co the presence of ^{68}Fe which, although associated to an extremely low production yield, was estimated to merit the long measurement needed for half-life determination. But the longer the measurements, the more difficult it is to control. No reference line could be used to monitor fragment production and isotopic separation. Three series of measurements were performed with similar E and q values (Table II).

^{68}Co

The ^{68}Co time correlations were first analyzed since the Co fragments are more abundant by a factor of 6 than the Fe ones and the half-life of the ^{68}Ni descendant is known (9) and long (19s) as compared with the expected value for ^{68}Co . Therefore, the only parameter to determine was the ^{68}Co half-life. Since, the ε and b values had not changed much from one measurement to

the other, values weighted by the number of fragments were used in a global analysis.

Given the low statistic, the M.L.H. method applied to all β's counted in a time t_c was found to be more appropriate. A half-life of 0.18 ± 0.10s was obtained for ^{68}Co. The large error bar accounts for the inaccuracy in ε and b (Table II and III).

^{68}Fe

In 200 hours of measurement, only 29 events of ^{68}Fe could be identified. Introducing the value obtained above as the half-life of the ^{68}Co descendant, the time correlations have been analyzed to extract the half-life of ^{68}Fe in a one parameter search using the M.L.H. procedure. A value of 0.10 ± 0.6 s has been obtained.

The ^{68}Fe nucleus is the source of a chain of 4 b's, schematically represented in fig. 3, the first two of which can be separated from background since they occur in shorter times than the background meantime.

Applying this criteria, a number of 5 possible β chains have been selected from the sequences following the 29 events of ^{68}Fe (Table IV). The number 5 is compatible with the probability ε^2 for detecting both β particles of the first two decays. Since the half-live of the descendant is much longer, the third column, $t_3 - t_0$, would correspond to a background occurence time. It gives indeed a meantime compatible with the background rate.

Assuming that these 5 sequences are due to β of the chains, and that there is no background counted between the parent fragment and the second β, the mean time values are evaluated to $< t > = (128 \pm 33)$ ms for the ^{68}Fe decay and to (316 ± 86) ms for the ^{68}Co one. The halflives expressed as $T = \ln 2 < t >$, are found to be (90 ± 23) ms and (221 ± 60) ms respectively, in agreement with the values obtained just above. The error bars are smaller because of the truncation in probabilities introduced by assuming that both β'

s of the chain are exclusively detected. Actually these sequences have only a finite probability of being associated to consecutive β decays.

Comparison with the Calculated Halflives

The halflives of n-rich Ni isotopes were well reproduced by the revised gross theory of Tashibana (10) and the microscopic calculations of Klapdor (11). But the half-lives of n-rich Co isotopes are calculated to be too large by a factor of 5 with both calculations. The values for ^{68}Co and ^{69}Co are found close to each other, like in our measurements. Recent microscopic calculations using the QRPA aproximation (12) have improved the order of magnitude of both halflives, but the value for ^{68}Co is predicted as four times larger than that of ^{69}Co , which is not compatible with our result.

The ^{68}Fe half-life provides a more stringent test of models than the two Co isotopes since it stands far from the last known half-life of an Fe isotope, ^{64}Fe. The values obtained by the first two calculations are larger than our result by a factor of 4, similar to the factor obtained for the two Cobalt isotopes. The recent calculation (12), however, gives a value compatible with our result.

Conclusion

Three more very neutron rich isotopes of Co and Fe have been identified in thermal fission of ^{239}Pu. The neutron excess reaches 10 units as compared to the most neutron rich stable isotope of Fe (^{58}Fe) as for the Ni isotopes previously reported (3). Given the large thermal fission cross-section of ^{239}Pu, the absolute production rates are large ≈ 50 μbarn for A=68, but the Lohengrin spectrometer is not primarily designed for exotic isotope studies. Its unique selection capacity of the rare isotopes - thanks to the combination with the ionization chamber - is combined with a low transmission coefficient of 10^{-6}. Further, the selection of a 1 MeV energy window in the distribution of

kinetic energy of the fragments rejects another factor of 10. Thus the present counting rates are very low.

Even with this limited accuracy, the half-lives reported here provide a guide for further calculations or extrapolations since the isotopes stand far from stability. On the chart of nuclei, they are located close to the limits of the area flooded by the high flux of neutrons in the process of rapid capture of neutrons occuring in nucleosynthesis (r-process). Among other properties, their β-decay halflives are required for an understanding of solar mass abundancies. Those half-lives are presently found shorter than they were in previous extrapolations thus, those isotopes will be less effective in the process since they decay too rapidly. The location and elemental composition of the first mass abundancy peak, close to A=80 should be reinterpreted.

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Table Captions

I Chain and isotopic production yields presently evaluated for (n, ^{239}Pu) thermal fission. The ^{235}U mass yields are indicated for comparison.

II Experimental conditions for the half-lives measurements.

III Results of the half-life evaluations of ^{69}Co, ^{68}Co and ^{68}Fe isotopes given in seconds.

IV Times sequences of 5 possible β chains emitted by ^{68}Fe fragments.

V Values of the half-lives calculated according to models referred to in the text and measured.values

Figure Captions

Fig. 1 : Energy loss spectra of the fragments selected in A/q by the spectrometer Lohengrin ; The measurement of the energy E_{Loh} gives the ionic state q and thus the value of A.

Fig. 2 : Times correlations of the three new isotopes of :

——— background

- - - - background with filiation

— — — Calculated with the indicated value of $T_{1/2}$

Fig. 3 : Scheme of the chain of the β's emitted by the ^{68}Fe fragments. The first β's can be identified since they are detected shortly after the fragments as compared with the background mean time.

A	Yield/fission ^{235}U	Yield/fission ^{239}Pu	Evaluated from q	Isotopic percentage Z=26	Z=27	Z=28	Z=29	Z=30
68		$6.6 \pm 1.3\ 10^{-8}$	17	2.6	11	80	6.5	
69	$2.2\ 10^{-8}$	$11 \pm 2\ 10^{-8}$	17 18		20 10	79 90		
73	$13\ 10^{-7}$	$15 \pm 3\ 10^{-7}$	18			3.8	55	41.2
82	$32\ 10^{-4}$	$19\ 10^{-4}$	→ From ref. 7 and 4 respectively					

Table I

Isotope	E	q	Rate of all fragments h^{-1}	Isotope rate h^{-1}	β rate s^{-1}	ε	Practibility ελ/b
^{69}Co	90 90	18 17	$1.5\ 10^{3}$ $1.6\ 10^{3}$	0.82 1.52	0.2 0.31	0.45 0.40	5.6 3.6
A=68	90	18	$3.8\ 10^{3}$	Co 0.83 Fe 0.15	0.42	0.35	3.2 5.8

Table II

Isotope	Number of fragments	First beta		All betas		Adopted value S_1
		χ^2	MLH	χ^2	MLH	
^{69}Co	175	0.29±0.06	0.31±0.07	0.24±0.05	0.26±0.05	0.27±0.05
^{68}Co	80				0.18±0.10	0.18±0.10
^{68}Fe	29				0.10±0.06	0.10±0.06

Table III

Detector number	t_1-t_0 (ms)	t_2-t_1 (ms)	t_3-t_0 (ms)	ΔE_β(keV)
6	170			135
		600		112
			1250	281
6	10			989
		340		223
			1380	127
3	200			186
		340		110
			2680	226
6	150			525
		80		232
			4610	118
4	110			272
		220		296
			1410	433

Table IV

Isotope	Calculated Half-lives (s)			Measured Halflives (s)
	Revised Gross Theory (10)	Microcopic Theories (11)	(12)	
^{69}Co	0.732	0.684	0.071	0.27±0.05
^{68}Co	0.798	0.813	0.290	0.18±0.10
^{68}Fe	0.370	0.419	0.160	0.10±0,06

Table V

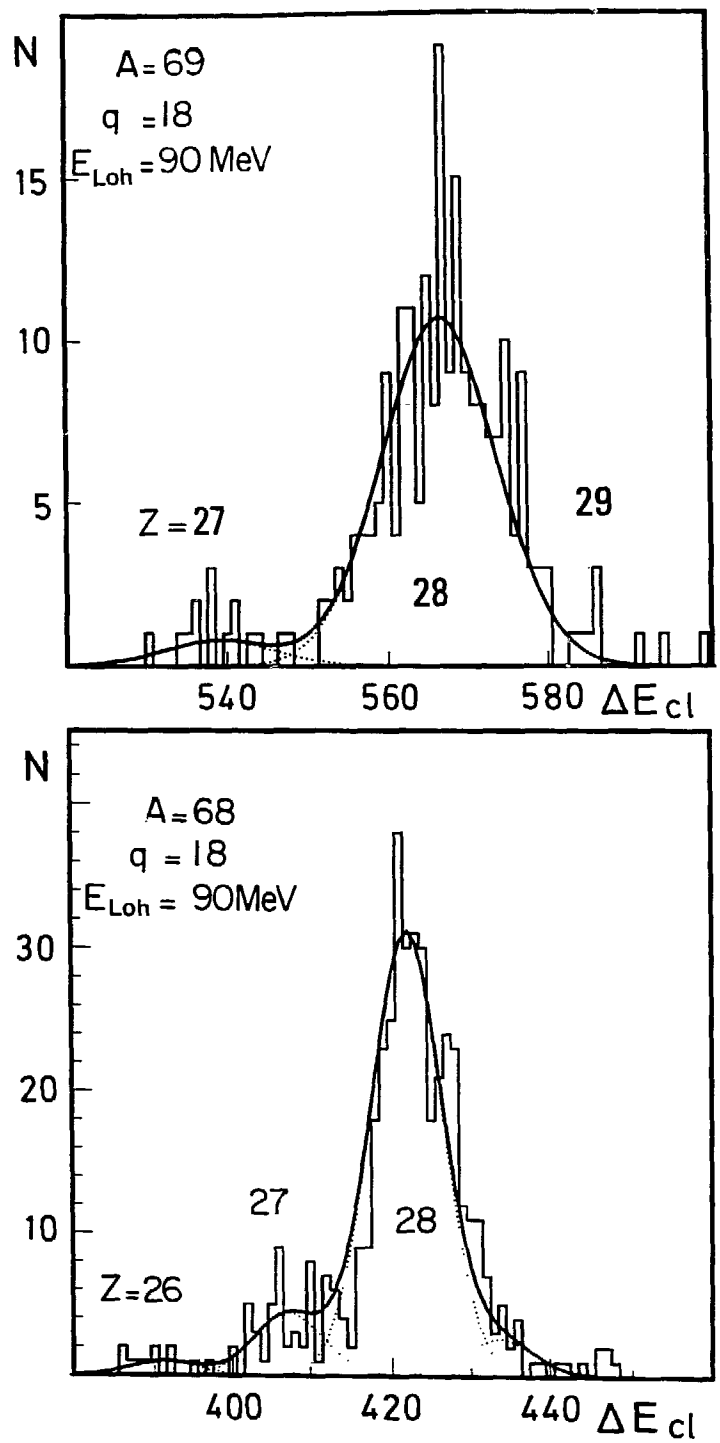
N
A=69
q =18
E_Loh =90 MeV
15
10
5
Z=27
28
29
540
560
580
ΔE cl
N
A=68
q =18
E_Loh = 90MeV
30
20
10
27
28
Z=26
400
420
440
ΔE cl

Fig 1

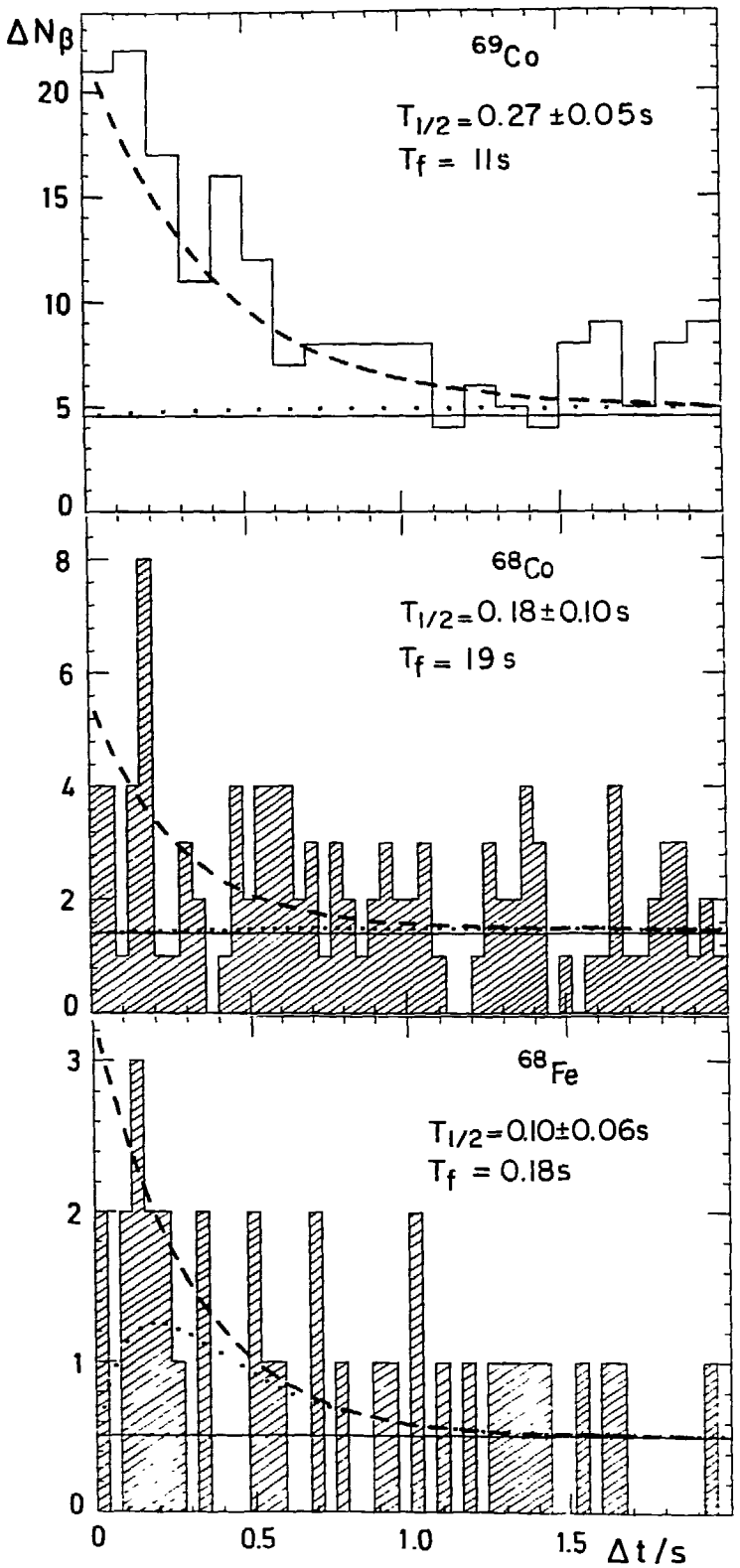
ΔNβ
69Co
T1/2 = 0.27 ±0.05s
Tf = 11s
68Co
T1/2 = 0.18±0.10s
Tf = 19 s
68Fe
T1/2 = 0.10±0.06s
Tf = 0.18s
Δt/s

Fig 2

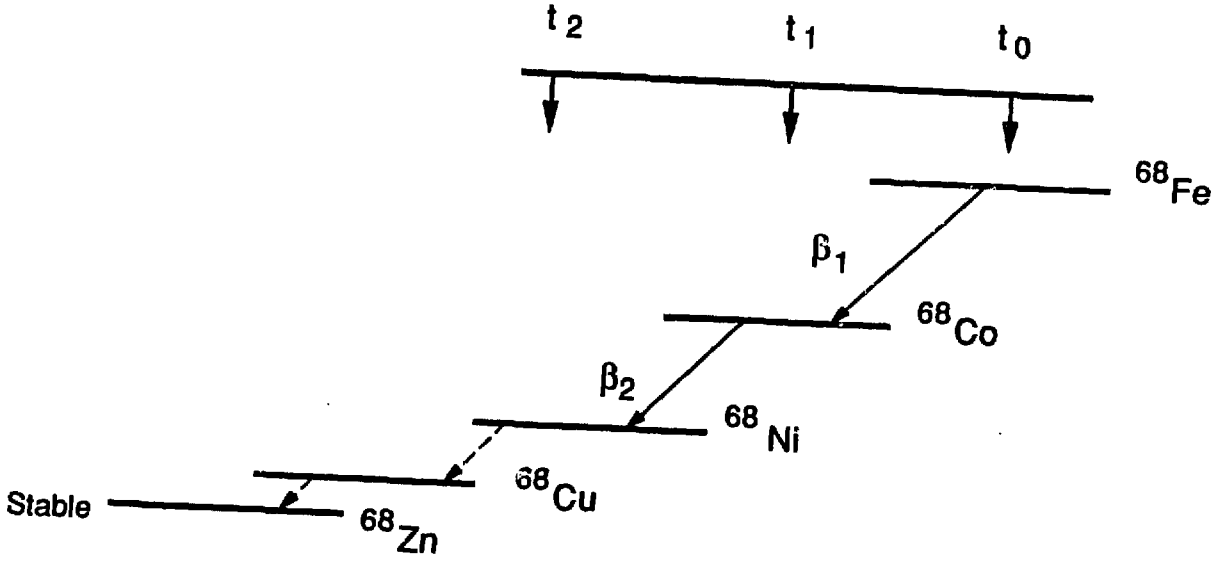

Fig 3