:GANIZATION FOR NUCLEAR RESEARCH

THERMAL NEUTRON INDUCED CHARGED PARTICLE REACTIONS IN RADIOACTIVE TARGETS OF ^{37}Ar, ^{109}Cd, ^{125}Xe, ^{127}Xe, AND ^{132}Cs

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ABSTRACT

Thermal neutron induced charged particle reactions in a radioactive target of ^{37}Ar have been studied. Upper limits of the cross-sections for the (n,α) reaction in radioactive targets of ^{109}Cd, ^{125,127}Xe, and ^{132}Cs have been obtained. The isotopically pure targets were produced at the ISOLDE facility at CERN and irradiated with thermal neutrons at the high flux reactor of the Institute Laue-Langevin in Grenoble. Charged particles from (n,p) and (n,α) reactions in ^{37}Ar were observed with cross-sections of 69 ± 14 b and 1970 ± 330 b, respectively. The Q-values for these reactions were determined to be 1600 ± 12 keV and 4630 ± 7 keV, in agreement with existing mass data. The branching ratio Γ_α/Γ_p of the ^{37}Ar capturing state was found to be 28.5 ± 2.7. An upper limit of the cross-section for the (n,γα) reaction in ^{37}Ar was obtained.

Nuclear reactions ^{37}Ar (n,p); ^{37}Ar, ^{109}Cd, ^{125,127}Xe, ^{132}Cs (n,α); E_n = thermal; measured Q_p, Q_α, Γ_α/Γ_p, deduced $\sigma_{n,p}$, $\sigma_{n,\alpha}$. Isotopically pure targets. Si surface barrier detector.

Geneva - 22 March 1978

(Submitted to Zeitschrift für Physik)

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1. INTRODUCTION

The last couple of decades have brought forward a number of detailed studies$^{1)}$ of proton and alpha spectra following the capture of thermal neutrons in heavy nuclei. Before this period, only a few (n_{th},p) and (n_{th},α) reactions had been studied, mainly in light nuclei, whereas by now several cases are known$^{1)}$ throughout the chart of nuclides. Many of the recent experiments on (n_{th},α) reactions have been performed at the Grenoble high-flux reactor, where a systematic study of this reaction in stable nuclides is being carried out$^{2-4)}$. In order to extend these investigations, we have undertaken a program to study the (n_{th},p) and (n_{th},α) reactions in neutron-deficient radioactive nuclides$^{5,6)}$ with half-lives longer than 10 hours. The recent developments in target and ion source techniques$^{7)}$ at the ISOLDE facility at CERN have made it possible to produce sufficient amounts of radioactive target material to make studies of these reactions feasible. When using neutron-deficient radioactive targets, which are characterized by large neutron separation energies and small proton and alpha separation energies, particle de-excitation of the capturing state will compete favourably with gamma de-excitation because of the large energy available for particle emission. These reactions are therefore expected to be more frequently encountered among neutron-deficient nuclides than among stable ones. If the Q-value for alpha emission is high enough and a large (n_{th},α) cross-section is found, there might also be a possibility to study the extremely rare two-step $(n,\gamma\alpha)$ reaction$^{8-10)}$.

In this paper we report the results obtained from neutron irradiations of radioactive targets of ^{37}Ar, ^{109}Cd, ^{125,127}Xe, and ^{132}Cs.

2. EXPERIMENTAL TECHNIQUES

Radioactive nuclides were produced in spallation reactions by bombarding a target with 600 MeV protons from the CERN Synchro-cyclotron. Most of the elements produced diffused out of the hot target into the ion source. Details on the different target-ion source systems used can be found in Table 1 and in the references given therein$^{11-13)}$.

The beam extracted from the ion source was separated into its constituent masses by the ISOLDE isotope separator[7]. The outcoming beam corresponding to the mass of interest was intercepted by a 10 μm thick foil of ultra-pure aluminium, and the 60 keV ions were implanted into a 0.2 cm^2 area of the foil. The neutron irradiations were performed at the end of a 87 m curved neutron guide (10^9 neutrons/$cm^2 \cdot s$) at the ILL high-flux reactor. The target position was viewed by a 450 mm^2, 100 μm thick silicon surface-barrier detector placed 45 mm from the target, outside the neutron beam. The detector had a resolution of 35 keV and subtended a solid angle of 2% of 4π sr.

The energy calibration was made with alpha particles from sources of ^{241}Am (5.486 MeV) and ^{232}Th (5.786 and 8.785 MeV). In addition to these standard sources the alpha peak from the $^6Li(n_{th},\alpha)^3H$ reaction (2.056 MeV) was also used. The energies, found for the proton and alpha peaks present in the ^{37}Ar spectrum (Fig. 1), were corrected for the different energy losses of protons and alpha particles in the detector gold layer and in the aluminium foil. The electronic stopping power tables by Northcliffe and Schilling[14] were used for this correction. The centroids of the strong peaks found in Fig. 1 were checked for a possible energy shift during the run. Any such shift was found to be less than 1 keV.

The number of target atoms (Table 2) for all but one of the five radioactive targets was determined by absolute counting of gamma-rays present in their decay. The exception was ^{37}Ar, which decays by pure EC [15] to the ground state of the stable nuclide ^{37}Cl, so the strongest detectable radiation is chlorine X-rays. Because of the low energy of the K X-rays, 2.62 keV, the ^{37}Ar source was placed inside a vacuum chamber, in front of a 200 mm^2 intrinsic germanium detector with an energy resolution of 240 eV at 5.2 keV. The efficiency of this detector at the Cl X-ray energy was determined by extrapolation from the points obtained with an open source of ^{54}Mn (K X-rays, 5.41, 6.00 keV) and the ^{109}Cd source (L X-rays, 3.02 keV). From an X-ray spectrum of the ^{37}Ar target recorded during one hour, the number of ^{37}Ar atoms at the start of the neutron irradiation was determined to be $(1.98 \pm 0.30) \times 10^{13}$.

3. EXPERIMENTAL RESULTS

3.1 Irradiation of 34.8 d ^{37}Ar

The charged particle spectrum obtained during a 139 h bombardment of the ^{37}Ar target is shown in Fig. 1. A prominent feature of this spectrum is the large peak at 4.14 MeV, which we assign to alpha particles from the $^{37}\mathrm{Ar}(n_{th},\alpha)^{34}\mathrm{S}$ reaction. The other strong peaks in the spectrum are due to (n_{th},α) reactions in ^{6}Li and ^{10}B present as impurities in the aluminium foil and in the walls surrounding the detector. Since the impurities are homogeneously distributed in these materials their corresponding alpha peaks are broadened and have large low-energy tails.

The energy of the alpha particles from the $^{37}\mathrm{Ar}(n_{th},\alpha)^{34}\mathrm{S}$ reaction was determined to be 4143 ± 6 keV. The corresponding Q_α value of 4630 ± 7 keV is in excellent agreement with the value 4629.8 ± 0.6 keV taken from the 1977 Mass Evaluation 16). In addition to this alpha branch, de-excitation of the capturing state by alpha emission to the 2.217 MeV first excited state in ^{34}S 15) is also energetically allowed and would contribute alpha particles with an energy of 2.24 MeV. However, no well-defined peak was found at this energy in the spectrum, and the upper intensity limit of this alpha branch was determined to be 0.3% of the ground-state branch.

A small peak with an energy of 1558 ± 12 keV and a FWHM of 32 keV was found between the α_0 and α_1 background peaks from the $^{10}\mathrm{B}(n_{th},\alpha)^{7}\mathrm{Li}$ reaction. This region of the ^{37}Ar spectrum, after subtraction of an exponential background, is shown in the inset of Fig. 1. We assign this peak as being due to protons from the $^{37}\mathrm{Ar}(n_{th},p)^{37}\mathrm{Cl}$ reaction, since the corresponding Q_p value 1600 ± 12 keV is in excellent agreement with the value 1597 ± 1 keV found by Parks et al. 17) for the inverse reaction.

From the contents of the proton peak, 4180 ± 400 counts, and of the alpha peak, 119460 ± 350 counts, the ratio of the alpha to proton widths for the ^{38}Ar capturing state was determined to be $\Gamma_\alpha/\Gamma_p = 28.5 \pm 2.7$.

The calibration of the neutron flux was made with a source of LiF, using the well-known cross-section of 941 ± 4 b for the $^{6}Li(n_{th},\alpha)^{3}H$ reaction[1]. The calibration source, containing $(2.33 \pm 0.18) \times 10^{16}$ ^{6}Li atoms, gave rise to a count rate of 150.0 ± 2.7 tritons per second when inserted in the neutron beam.

The cross-sections for the (n_{th},α) and (n_{th},p) reactions in ^{37}Ar were determined to be 1970 ± 330 b and 69 ± 14 b, respectively.

3.2 Irradiations of 453 d ^{109}Cd, 16.8 h ^{125}Xe, 36.4 d ^{127}Xe, and 6.5 d ^{132}Cs

For all these nuclides the charged particle spectra recorded during the neutron irradiations showed no apparent peaks at the energies expected[16] for alpha particles from (n_{th},α) reactions. Only upper limits for the reaction cross-sections were obtained. All relevant information on these experiments is summarized in Table 2. Preliminary results from the ^{127}Xe and ^{132}Cs irradiations have previously been published[6].

4. DISCUSSION

With the large average level spacing [calculated[18] to be 8 keV at the neutron separation energy[16] 11.84 MeV] in the N = 20 nucleus ^{38}Ar, one expects that only one excited state, close to the neutron separation energy, will contribute significantly to the capture cross-section. The ground-state spin and parity[15] of ^{37}Ar is $3/2^{+}$, and the capturing state in ^{38}Ar is consequently either 1^{+} or 2^{+} (Fig. 2). With the very large cross-section found for the (n_{th},α) reaction a 1^{+} assignment is ruled out, since alpha emission from a 1^{+} state to the 0^{+} ground state of ^{34}S is parity forbidden. We thus assign the ^{38}Ar capturing state spin and parity as 2^{+}.

In the ^{37}Ar spectrum (Fig. 1) there is no sign of the broad hump which is characteristic of the $(n,\gamma\alpha)$ reaction. The upper limit of the cross-section for this reaction was determined to be 5 barns. Statistical model calculations of the average partial widths for 2^{+} states in the region of the ^{38}Ar capturing state yield Γ_{α} = 160 eV, Γ_{p} = 22 eV, and Γ_{γ} = 1.2 eV, indicating that alpha emission is the dominating decay mode for this state. Very little strength is then left to the primary gamma-rays in the $(n,\gamma\alpha)$ process, making this process highly unfavourable despite the large (n_{th},α) cross-section.

The ^{37}Ar absorption cross-section for thermal neutrons would be 2040 barns if gamma de-excitation of the capturing state in ^{38}Ar is very small or non-existent. If not, the cross-section would be even larger. The large ^{37}Ar neutron cross-section cannot be due to neutron capture into a far-off tail of a resonance in ^{38}Ar, but is more likely coming from a region close to the centre of a resonance. We therefore expect a 2^+ state to lie very close to the neutron separation energy in ^{38}Ar.

We are grateful to the Institute Laue-Langevin and to the ISOLDE Collaboration for the facilities put at our disposal, and to O.C. Jonsson and R. Deltenre for the handling of the radioactive sources.

Table 1

Element produced	Target			Proton beam (μA)	Refs.
	Material	Thickness (g/cm^2)	Temperature (°C)		
Ar	VC	38	2100	1.0	11
Cd	Sn	115	1200	1.6	12
Cs [a)]	La	140	1100	0.7	13

a) ^{125,127}Xe were obtained as the daughters of ^{125,127}Cs.

Table 2

Nuclide	Half-life	Irradiation time (h)	Amount of target material (atoms)	Reaction	Q-value (MeV)	Cross-section (b)	Refs.
^{37}Ar	34.8 d	139	1.98×10^{13}	(n_{th},p)	1.60	69 ± 14	This work
				(n_{th},α)	4.63	1970 ± 330	" "
				$(n_{th},\gamma\alpha)$		≤ 5	" "
^{109}Cd	453 d	190	1.54×10^{14}	(n_{th},α)	7.02	≤ 0.05	" "
^{125}Xe	16.8 h	47	1.4×10^{15}	(n_{th},α)	8.85	≤ 0.03	" "
^{127}Xe	36.4 d	170	8.2×10^{14}	(n_{th},α)	7.85	≤ 0.01	6
^{132}Cs	6.5 d	130	4.0×10^{14}	(n_{th},α)	6.98	≤ 0.15	6

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Figure captions

Fig. 1 : Energy spectrum of charged particles from a 120 µCi target of ^{37}Ar irradiated with thermal neutrons (flux 10^9 cm^{-2} s^{-1}) for 139 h. Towards the low-energy part of the spectrum there is a significant increase in the number of counts due to a background originating from scattered neutrons. Superimposed on this background are some peaks due to (n_{th},α) reactions in ^{6}Li and ^{10}B present as impurities. The peak at 4.143 MeV has been identified as due to alphas from the $^{37}Ar(n_{th},\alpha)^{34}S$ reaction. Subtracting an exponential background from the part of the spectrum shown in the small quadrangle results in the spectrum shown in the inset. The peak at 1.558 MeV is identified as protons from the $^{37}Ar(n_{th},p)^{37}Cl$ reaction.

Fig. 2 : Level scheme for the (n_{th},α) and (n_{th},p) reactions in ^{37}Ar.

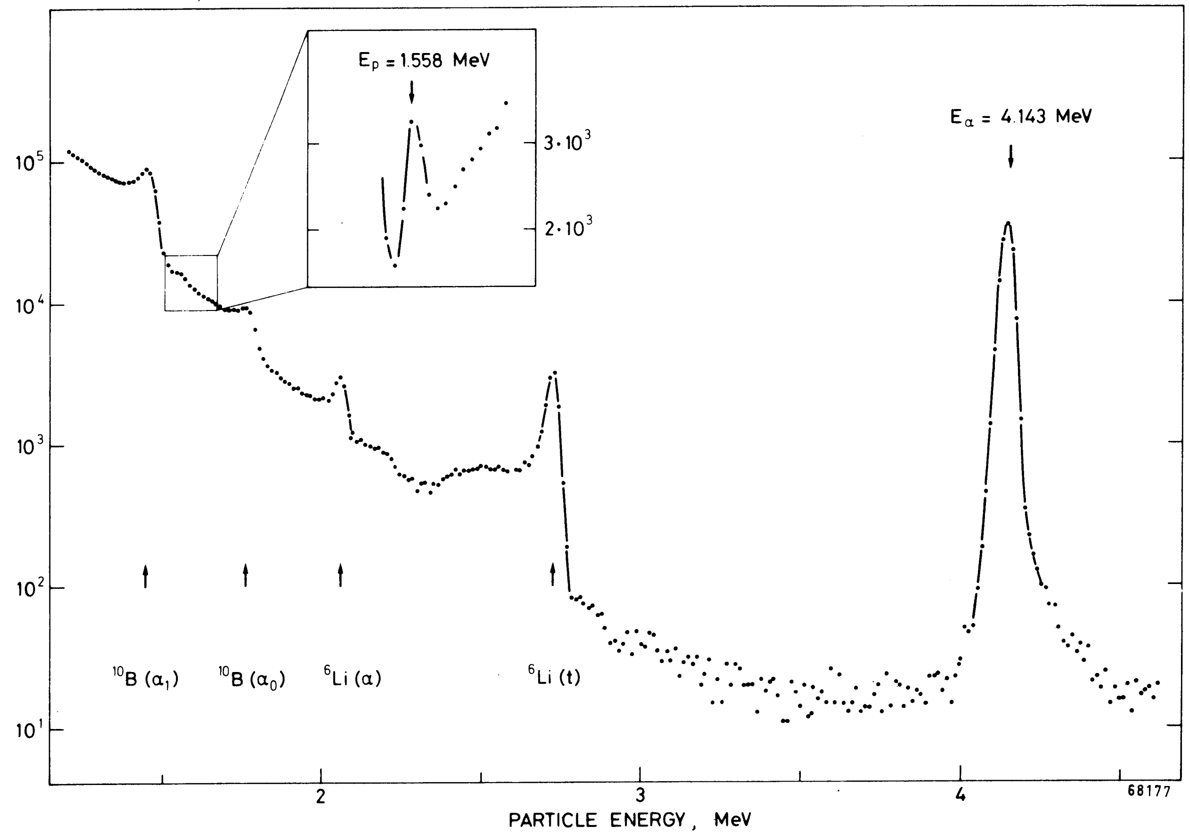
E$_p$ = 1.558 MeV
3·10^3
2·10^3
E$_\alpha$ = 4.143 MeV
10^5
10^4
10^3
10^2
10^1
^{10}B (α_1)
^{10}B (α_0)
^{6}Li (α)
^{6}Li (t)
2
3
4
68177
PARTICLE ENERGY, MeV

Fig. 1

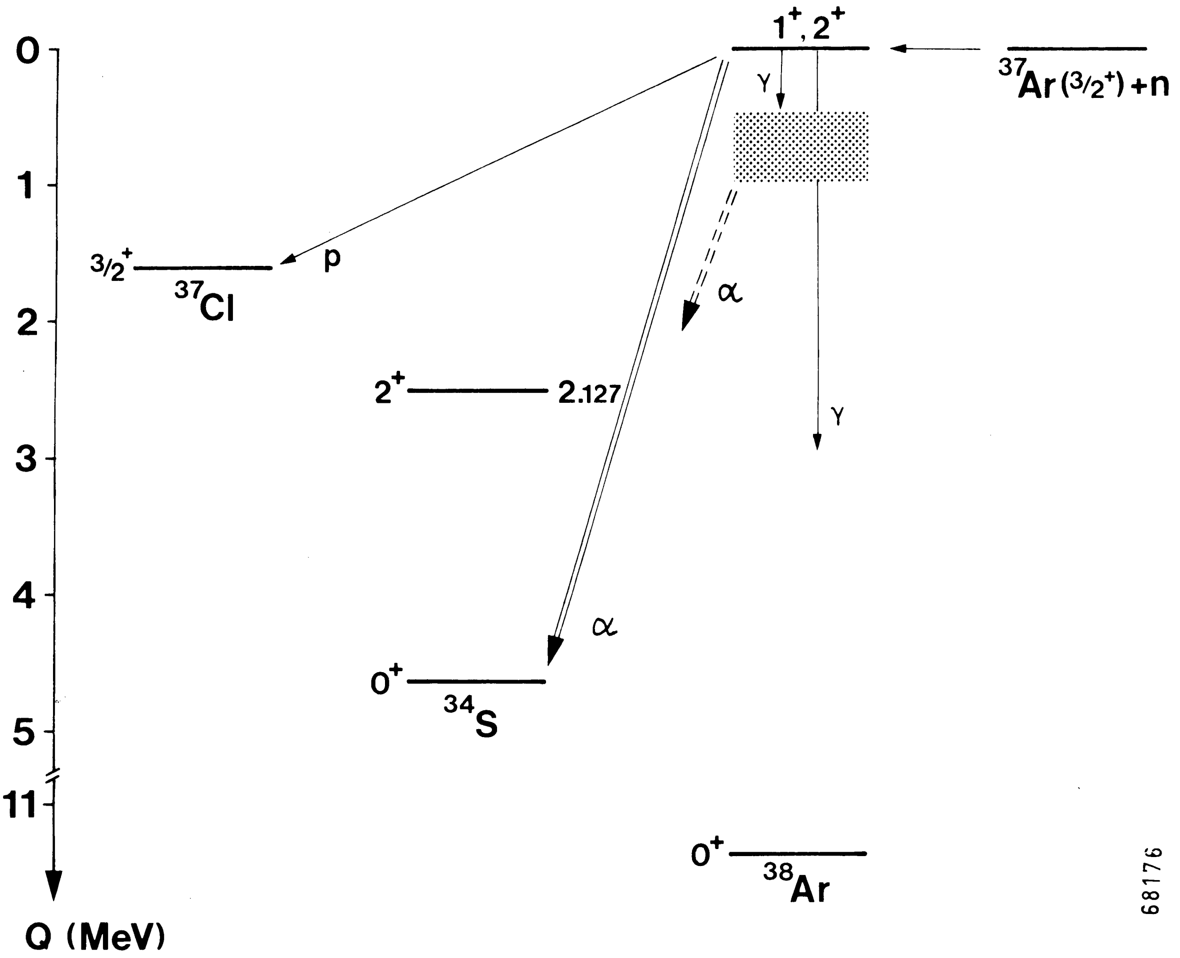
0
1
2
3
4
5
11
Q (MeV)
3/2+
37Cl
p
2+
2.127
0+
34S
α
α
γ
γ
1+, 2+
37Ar(3/2+)+n
0+
38Ar
68176

Fig. 2