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NEUTRON ACTIVATION CROSS SECTIONS ON Ti ISOTOPES AT 14.8 MeV

I. Ribanský and Š. Gmuca

Institute of Physics EPRC, Slovak Academy of Sciences,
Dúbravská cesta, CS-842 28 Bratislava, Czechoslovakia

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ABSTRACT

Neutron activation cross sections on Ti isotopes at 14.8 MeV were measured using the Ge(Li) γ-ray spectroscopy. The linear least squares method was used for resolving the interfering reactions on neighbouring isotopes. The results obtained are discussed in detail and are compared with preequilibrium, equilibrium and semiempirical calculations.

1. Introduction

The most recent updated edition of the WRENDA 81/82 (1981) issued by NDS IAEA contains a large number of requests for improvement of the 14 MeV neutron cross sections, required for the development of fission and fusion reactor technologies.

For measuring these cross sections, the activation method is the most widely used. Most of the experimental activation cross sections given in the literature display an appreciable margin of uncertainty; and the discrepancies between the values given by different authors exceed considerably the limits of experimental errors. A survey of published studies on the activation cross section measurements suggests, that these discrepancies might originate in the experimental technique.

Most of the earlier measurements have been made by β-counting or by γ-counting with NaI(Tl) detectors. The drawback of β-counting is the difficulty in resolving the continuous β-spectrum into different half-life components. This gives rise to the unreliability of the results, especially when many residual activities are involved in one sample, or when a thick sample is used. In the case of γ-counting with NaI(Tl) detectors, difficulties arise in the identification of the characteristic γ-rays from the reaction products, due to a poor resolving power of the detector. These drawbacks are obviated with the use of a Ge(Li) detector. Its high resolution available for γ-ray identification makes it possible to obtain

reliable and accurate reaction cross sections even in the presence of a high background.

On the other hand, there are factors (usually not taken into account) which may seriously affect the results of measurement even when the Ge(Li) detector is used. Cases exist where interfering reactions on impurities, or neighbouring isotopes leading to the same reaction product, exert a substantial influence on the cross section measured. Great care must be taken when, due to a low induced activity, a close sample-detector counting geometry is used. Coincidence summing effects on cascading γ-rays may easily become as high as several tens of percent in this case.

Under these circumstances it is difficult to draw meaningful conclusions from comparisons between the predictions derived on the basis of various models, and the experimental data obtained by measurement.

In the present work, we propose to measure the activation cross sections of Ti isotopes at 14.8 MeV using a Ge(Li) detector and to compare the results for (n,p) reactions with the predictions based on the preequilibrium model, the evaporation model and a semiempirical formula. Because of the strong competition between the (n,p) and (n,np) [a] reactions in this mass region, measurements on samples with different isotopic abundances have been performed, and the linear least squares method has been used

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a) The (n,np) reaction means the sum of (n,n'p), (n,pn) and (n,d) reactions

to resolve the cross section of the reactions leading to the same activation product. The coincidence summing corrections have been applied wherever needed.

2. Experimental procedure

2.1 Sample preparation

All samples used (except those of natural isotopic composition) were prepared by pressing TiO_2 powder into plexiglass containers (ø 16 mm). The enriched samples were supplied by TECHSNABEXPORT, Moscow (USSR). The samples of natural isotopic abundance were prepared from a thin metal foil. The isotopic abundances of the Ti samples used are listed in Table 1. All the samples were of spectral purity. No chemical impurities were found by means of the XRFA method and by checking the γ-spectra of the irradiated samples by means of a Ge(Li) detector. The sample thickness varied from 100 to 300 mg/cm^2; it was chosen by taking into account the magnitude of the expected cross sections, the corrections to be applied, and the neutron yield.

2.2 Neutron irradiation and fluence monitoring

The requisite neutrons were obtained from the T(d,n)^{4}He reaction using incident deuteron beams of about 120 keV from a small Cockcroft-Walton accelerator. The tritium target (ø 45 mm) consisted of 25 Ci of tritium absorbed in a Ti layer on a thin Cu plate with its back directly cooled by water. The geometry of irradiation was carefully checked. The sample - TiT target distance was 9 mm and the position and size of the beam spot were defined by a slit

(ø 6 mm) near the target. The neutron energy was 14.8 MeV and its spread was estimated as ± 250 keV (see Csikai 1981). The time variation of the neutron yield was monitored by two neutron detectors. One of them served for recording recoil protons from a thin polyethylene radiator with a CsI scintillator (thickness 1 mm) and the other for detecting thermalized neutrons using Li-glass. For the given irradiation geometry both monitors were calibrated several times before each run, using the $^{56}Fe(n,p)^{56}Mn$ reaction (Ryves et al 1978) which is considered to be a suitable secondary standard for fast neutron fluence monitoring.

2.3 γ-counting of irradiated samples

The γ-ray activities of the irradiated samples were measured with a 50 cm^3 close-ended coaxial Ge(Li) detector (resolution 2.5 keV at 1332 keV). The samples were put directly on the detector's cup, at its axis to preserve a rotational symmetry. The full-energy peak efficiency and the total efficiency of this detector at the given geometry were determined with an accuracy of 1.5 % and 5 %, respectively, as described by Gmuca and Ribanský (1982). Both efficiencies are shown in Fig. 1. The total efficiency is needed for a correct calculation of the coincidence summing corrections of the cascading γ-rays (Debertin and Schötzig 1979). This effect may seriously affect the areas of full energy peaks measured at a close geometry, and thus neglecting them may lead to erroneous results.

2.4 Data reduction

Our data acquisition and control system is described elsewhere (Gmuca and Ribanský 1983 a). Therefore, only the details concerning the present experiment will be given here.

The measured data, recorded in a disc file, were processed off-line on a TPA-70 minicomputer. The γ-ray spectra were analysed by the program GWENN (Gmuca and Ribanský 1983 b) based on a nonlinear least squares fit to the semiempirical peak shape function. The areas of the full-energy peaks were corrected for the coincidence summing effects, and for the selfabsorption of γ-radiation in the sample. The program KORSUM of Debertin and Schötzig (1979) was used for the calculation of the coincidence summing corrections. The accuracy of the efficiencies of the Ge(Li) detector, and that of the decay data of the reaction products (Lederer and Shirley 1978) are sufficient for calculating these corrections with an uncertainty well below 1 % in this case. The decay data of the reaction products observed are given in Table 2 together with the calculated corrections for the coincidence summing effects.

Since in the lighter mass region A≤60 the (n,np) process competes seriously with the (n,p) reaction, measurements were made on samples with several different isotopic abundances. The cross sections of reactions (n,p) and (n,np) on neighbouring isotopes leading to the same reaction product were, therefore, determined simultaneously and in a consistent way, by a linear least squares method.

As a rule, samples with three to five different isotopic abundances were used in this experiment (see Table 1)

3. Results and uncertainties

The final results of our measurements are listed in Table 3. For each group of reactions (n,p) and (n,np) leading to the same activation product, the correlation matrix resulting from the linear least squares decomposition is given. An attempt was made to distinguish between uncorrelated (random) errors and the correlated (systematic) ones. In order to simplify the decomposition of the competing reactions and to avoid the solution of a monstrous system of linear equations, the data entering the linear least squares analysis were considered to be mutually independent, taking into account the statistical errors of the full-energy peak areas only. The systematic errors were added in quadrature after resolving the yield of each reactione. The correlation matrix thus refers to the uncorrelated (statistical) part of the total error only. The main sources of systematic errors considered are listed in Table 4. All errors quoted represents one standard deviation.

4. Theoretical predictions of (n,p) cross sections

4.1 Preequilibrium model

The presence of the preequilibrium mechanism, in addition to the compound nucleus processes, were already demonstrated in neutron induced reactions (see e.g. Millazzo-Colli and Braga-Marcazzan 1973). The exciton model, as

originally proposed by Griffin (1966) and later modified and improved by a number of authors, is most widely used for the analysis of this part of the reaction. In the present work, the modified exciton model proposed by Gmuca and Ribanský (1980) has been used. This modification is based on the assumption that only the nucleons near the Fermi energy of the target nucleus take part in the pre-equilibrium stage of the nuclear reaction. This approach uses the density of n-exciton states and the density of final accessible states corrected for the finite effective depth of the hole excitations E_H instead of those for the infinite potential well as usually used. E_H may be estimated as (Gmuca 1982)

$$E_H = \sqrt{E/a} \ , \qquad (1)$$

where E is the excitation energy and a the level density parameter. For normalization of the results the averaged squared matrix element $|M|^2$ has to be known. For the calculation of $|M|^2$ the following expression was proposed (Gmuca 1982)

$$|M|^2 = K . E_H^{-1} . g^{-3}, \qquad (2)$$

which takes into account the finite depth of the hole excitations. g is the single particle state density which is related to the level density parameter a by the well known formula

$$g = \frac{6}{\pi^2} \cdot a \qquad (3)$$

The above expression for $|M|^2$ (2) leaves K as a free pa-

rameter. Recently, this parameter was estimated as 0.30 by the analysis of neutron and proton spectra emitted in neutron induced reactions (Gmuca 1982). Since in the present work the closed form of the modified exciton model was used the standard evaporation spectrum had to be added. The level density parameter a was taken from Gilbert and Cameron (1965). Examples of calculations with this model are shown in Figs. 2, 3 where the calculated primary proton spectra are compared with experimental data for (n,xp) reactions on ^{46,48}Ti measured by Grimes et al (1977) at 15 MeV. As one can see, the model is in fair agreement with the experimental data in high energy region where only the emission of one particle is possible. The emission of only the first particle is considered in the calculations since we are interested primarily in the preequilibrium emission; and it is known that the low-energy part of the experimental spectra may be well described by a statistical multiparticle emission (Grimes et al 1978). With this model the calculations of the (n,p) spectra for all Ti isotopes at 14.8 MeV were performed. For comparison with our measured, activation cross sections, the integral values of the primary proton spectra up to the threshold of the second particle emission were calculated. These values are considered as reasonable lower limits of the activation cross sections. The final results are given in Table 5.

4.2 Evaporation model

The cross sections of (n,p) reactions calculated from the pure evaporation model are also shown in Table 5. These values are based on a level density at energy E given by

$$\rho(E) = (E - \delta)^{-1} \exp\left\{2\left[a\,(E - \delta)\right]^{1/2}\right\}, \qquad (4)$$

where δ is the pairing correction taken from Nemirovsky and Adamchuk (1962) and a is the level density parameter given by Gilbert and Cameron (1965). The calculated values were obtained in the same manner as in the preceding paragraph.

4.3 Semi-empirical predictions

There exist several semi-empirical formulae predicting the (n,p) cross section at 14 MeV. A simple formula for the (n,p) cross section in the form

$$\sigma_{n,p} = 45.2\,(A^{1/3} + 1)^2 \exp\left\{-33(N - Z)/A\right\}\ \text{(mb)} \qquad (5)$$

derived by Levkovskii (1964) was used. This formula succesfully describes the experimental data in a wide range of nuclei $12 < A < 170$. This is quite surprising since for the heavier nuclei the preequilibrium mechanism of a proton emission is dominant while in the lighter region the compound nucleus mechanism seems to prevail. Thus this formula is able to describe both different reaction mechanisms. The (n,p) cross section values predicted from the above formula are also shown in Table 5.

5. Discussion

5.1 $^{46}Ti(n,p)^{46m,g}Sc$

Our isomeric cross section σ_m (55.0 ± 2.2 mb) can be directly compared with the result of 66 ± 16 mb by Jaczyszyn

et al (1973) obtained with a NaI(Tl) detector, and with that of 48±8 mb by Molla and Qaim (1977) obtained by Ge(Li) γ-spectroscopy. All results are consistent within the limits of errors quoted.

Since in most of the previous experiments the total (n,p) reaction cross section was measured, our ground-state cross section σ_g (211.7±8.3 mb) cannot be compared with other results.

The total (n,p) reaction cross section σ_{m+g} obtained in this work (266.7±8.6 mb) is in a good agreement with the ENDF/B-IV (1975) evaluation (253 mb) and our preequilibrium calculation. The equilibrium calculation seems to be rather high while Levkovskii's prediction is too low.

5.2 $^{47}Ti(n,p)^{47}Sc$

The measured value of this cross section (169.5±6.9 mb) is substantially higher than the ENDF/B-IV evaluation (108 mb), and well ontside the experimental uncertainties of most of the other measurements. Thus, our result is 56 % above the evaluation. In comparison with calculations, the equilibrium calculation and Levkovskii's prediction are nearly consistent with the evaluated cross section. The preequilibrium model calculation is much higher, but not so high as the result of our measurement. One must keep in mind, however, that the calculated data yield only the lower limits of the activation cross sections.

5.3 $^{48}Ti(n,p)^{48}Sc$

Our value of this cross section, 71.7±2.6 mb, is some-

what higher than the ENDF/B-IV evaluation (66.0 mb). However, in this case the coincidence summing effects (in our case requiring a correction of +23.7 %) may play an important role in some measurements entering the evaluation. Our experimental value agrees with the preequilibrium calculation while the equilibrium calculation and Levkovskii's prediction appear too low.

5.4 $^{50}Ti(n,p)^{50}Sc$

This reaction has not been included into the ENDF/B-IV Dosimetry File (1975). Our result agrees well with the preequilibrium calculation and Levkovskii's prediction. The equilibrium calculation is much lower, thus indicating an important role of the preequilibrium reaction mechanism in this case. Our cross section value is consistent with most of the previous activation measurements within the limits of their quoted errors.

It is known that the nucleus ^{50}Sc possesses a metastable state with a half-life of 0.35 s and an energy 256.9 keV decaying completely by an isomeric transition (Lederer and Shirley 1978). In our work the isomeric cross section has not been determined due to a short half-life. Hence, our experimental value refers to the total (n,p) activation cross section.

5.5 $^{47}Ti(n,np)^{46m,g}Sc$

The isomeric σ_m and ground-state σ_g cross section of this reaction have been determined for the first time (see Table 3). In the previous measurements the total

(n,np) reaction cross section was measured. Our value 62.8±2.4 mb of σ_{m+g} is about 46 % above the ENDF/B-IV evaluation value 43 mb which is, in fact, based only on one measurement of Pai (1966).

5.6 $^{48}Ti(n,np)^{47}Sc$

The cross section value given in Table 3 is somewhat lower than the ENDF/B-IV evaluation 14.0 mb based on the measurement by Pai (1966), and compares fairly well with 12.4±0.2 mb according to Cross (1963) and 9±2 mb according to Qaim (1982), quoting their estimate of uncertainty.

5.7 $^{49}Ti(n,np)^{48}Sc$

This reaction was not included into the ENDF/B-IV evaluation. Our cross section value 6.9±0.7 mb is consistent with 9±2 mb by Cross (1963), 9±2 mb by Pai (1966), and 7.8±3.0 mb by Qaim (1982) within the quoted limits of errors.

5.8 $^{50}Ti(n,\alpha)^{47}Ca$

Our cross section value 9.0±0.8 mb of this reaction is in excellent agreement with all other results: 10±5 mb by Hillman (1962), 8.6±0.2 mb by Cross (1963), 9.4±2.8 mb by Yu-Wen-Yu (1967) and 10±2 mb by Levkovskii et al (1969).

5.9 Preequilibrium fraction emission

In Section 4.1 our experimental (n,p) cross sections

on Ti isotopes were evaluated by means of the modified exciton model in combination with Weisskopf-Ewing evaporation model. An important quantity characterizing the competition between both different reaction mechanisms is the so called fraction of preequilibrium emission F_{PEQ}. This denotes the relative contribution of the preequilibrium emission to the respective reaction. In the case of the emission of protons, the preequilibrium fraction F^{P}_{PEQ} may be simply expressed as

$$F^{P}_{PEQ} = \sigma_{PEQ}(n,p)/\sigma(n,p) \ , \qquad (6)$$

where $\sigma_{PEQ}(n,p)$ is the preequilibrium part of the (n,p) reaction cross section and $\sigma(n,p)$ the total (n,p) reaction cross section. The final results of this type of analysis are also shown in Table 5, together with the experimental (n,p) cross sections and the calculated ones. One can see that the fraction of the preequilibrium proton emission exhibits a surprisingly strong isotopic dependence. While in the ^{46}Ti(n,p) reaction the preequilibrium emission takes only 7.2 % of the reaction cross section, in the ^{50}Ti(n,p) the preequilibrium fraction reaches as much as 46.4 %. This indicates that the preequilibrium reaction mechanism may play an important role in the fast neutron induced proton emission also in the lighter mass region, for the nuclei with a higher neutron excess.

6. Summary

The precise neutron activation cross sections of Ti isotopes at 14.8 MeV were measured in consistent way by

means of Ge(Li) γ-spectroscopy. The linear least squares method was used to resolve the (n,p) and (n,np) reactions on the neighbouring isotopes leading to the same activation product. The obtained data were compared with the ENDF/B-IV evaluations wherever possible. An evident disagreement was found in the case of ^{47}Ti(n,p) and ^{47}Ti(n,np) reactions. Apparently, additional measurements are needed for these reactions to remove the discrepancies.

The (n,p) cross sections have also been compared with calculations based on preequilibrium and equilibrium models and on Levkovskii's predictions. The evaporation calculation, however, fails with increasing neutron number, while Levkovskii's formula fails with decreasing neutron number in the titanium isotopic chain. It seems that the only model able to describe the isotopic dependence of the (n,p) reaction cross section correctly is the modified exciton model which, in addition, describes well also the primary proton spectra from the reactions ^{46}Ti(n,xp) and ^{48}Ti(n,xp) (see Figs. 2,3) as measured by Grimes et al (1977). An interesting feature was drawn from this analysis: the fraction of the preequilibrium proton emission seems to exhibit a strong isotopic dependence. It appears worthwile to study this interesting behaviour in a greater detail. Such investigation is currently in progress.

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TABLE 1: Isotopic abundances (%) of samples

Sample	Isotope				
	46	47	48	49	50
Natural Ti	8.2	7.4	73.8	5.4	5.2
^{46}Ti	73.8	2.6	20.1	1.7	1.8
^{47}Ti	2.5	76.1	19.0	1.3	1.1
^{48}Ti	0.2	1.0	97.8	0.7	0.3
^{49}Ti	3.3	3.5	22.3	67.1	3.8

TABLE 2: Decay data of reaction products

Reaction product	$T_{1/2}$	E_γ (keV)	I_γ (%)	Coincidence summing corrections
^{46m}Sc	18.7 s	142.5	62.1	1
^{46g}Sc	83.8 d	889.2	100	1.109
		1120.5	100	1.108
^{47}Sc	3.42 d	159.4	68.5	1
^{48}Sc	43.7 h	983.5	100	1.237
		1037.5	97.5	1.237
		1212.8	2.38	1.022
		1312.1	100	1.237
^{50}Sc	1.71 m	523.7	88	1.228
		1121.0	100	1.211
		1553.7	100	1.211
^{47}Ca	4.536 d	1297.1	77.2	0.997

TABLE 3: Activation cross sections with 14.8 MeV neutrons of Ti isotopes from the present work

Reaction	Measured cross section (mb)	Relative error total (%)	Relative error uncorrel. (%)	Correlation matrix (%)	
$^{46}Ti(n,p)^{46m}Sc$	55.0±2.2	4.0	0.6	100	
$^{47}Ti(n,np)^{46m}Sc$	6.92±0.33	4.7	2.5	-45	100
$^{46}Ti(n,p)^{46g}Sc$	211.7±8.3	3.9	1.6	100	
$^{47}Ti(n,np)^{46g}Sc$	55.9±2.3	4.1	2.2	-14	100
$^{46}Ti(n,p)^{46m+g}Sc$	266.7±8.6	3.2			
$^{47}Ti(n,np)^{46m+g}Sc$	62.8±2.4	3.8			
$^{47}Ti(n,p)^{47}Sc$	169.5±6.9	4.1	1.1	100	
$^{48}Ti(n,np)^{47}Sc$	11.52±0.51	4.4	1.8	-51	100
$^{48}Ti(n,p)^{48}Sc$	71.7±2.6	3.7	0.5	100	
$^{49}Ti(n,np)^{48}Sc$	6.9±0.7	10.1	9.5	-28	100
$^{50}Ti(n,p)^{50}Sc$	15.40±0.63	4.1	1.4	-	
$^{50}Ti(n,\ )^{47}Ca$	9.0±0.8	8.9	8.6	-	

TABLE 4: The principal sources of correlated uncertainty

Source of error	Resulting uncertainty
Calibration of monitors including reference reaction $^{56}Fe(n,p)$	3.6 %
Efficiency of Ge(Li) detector	1.5 %
Coincidence summing corrections	0.7 %
Sample composition (chemical, isotopic)	0.2 %
Mass	0.1 %
Irradiation and counting geometry	0.2 %

TABLE 5: Comparison between experimental (n,p) cross sections and various predictions

Reactions	Exp. cross section (mb)	Calc. cross sections (mb) (1)	(2)	(3)	F^{P}_{PEQ}
$^{46}Ti(n,p)$	266.7±8.6	259	285	229	0.072
$^{47}Ti(n,p)$	169.5±6.9	141	127	115	0.180
$^{48}Ti(n,p)$	71.7±2.6	69	58	58	0.237
$^{50}Ti(n,p)$	15.40±0.63	14.7	10.8	15	0.464

(1) - Preequilibrium model

(2) - Evaporation model

(3) - Levkovskii formula

Figure captions

Fig. 1: The absolute full energy peak efficiency (FEPE) and total efficiency (TE) of our Ge(Li) detector at close geometry.

Fig. 2: The comparison between the experimental data (histogram) of Grimes et al (1977) and the theoretical proton spectrum calculated by the modified exciton model (solid line) for the $^{46}Ti(n,p)$ reaction. The dashed line represents the preequilibrium part of the proton emission. The arrow indicates the threshold for the second particle emission.

Fig. 3: The same as in Fig. 2 for the $^{48}Ti(n,p)$ reaction.

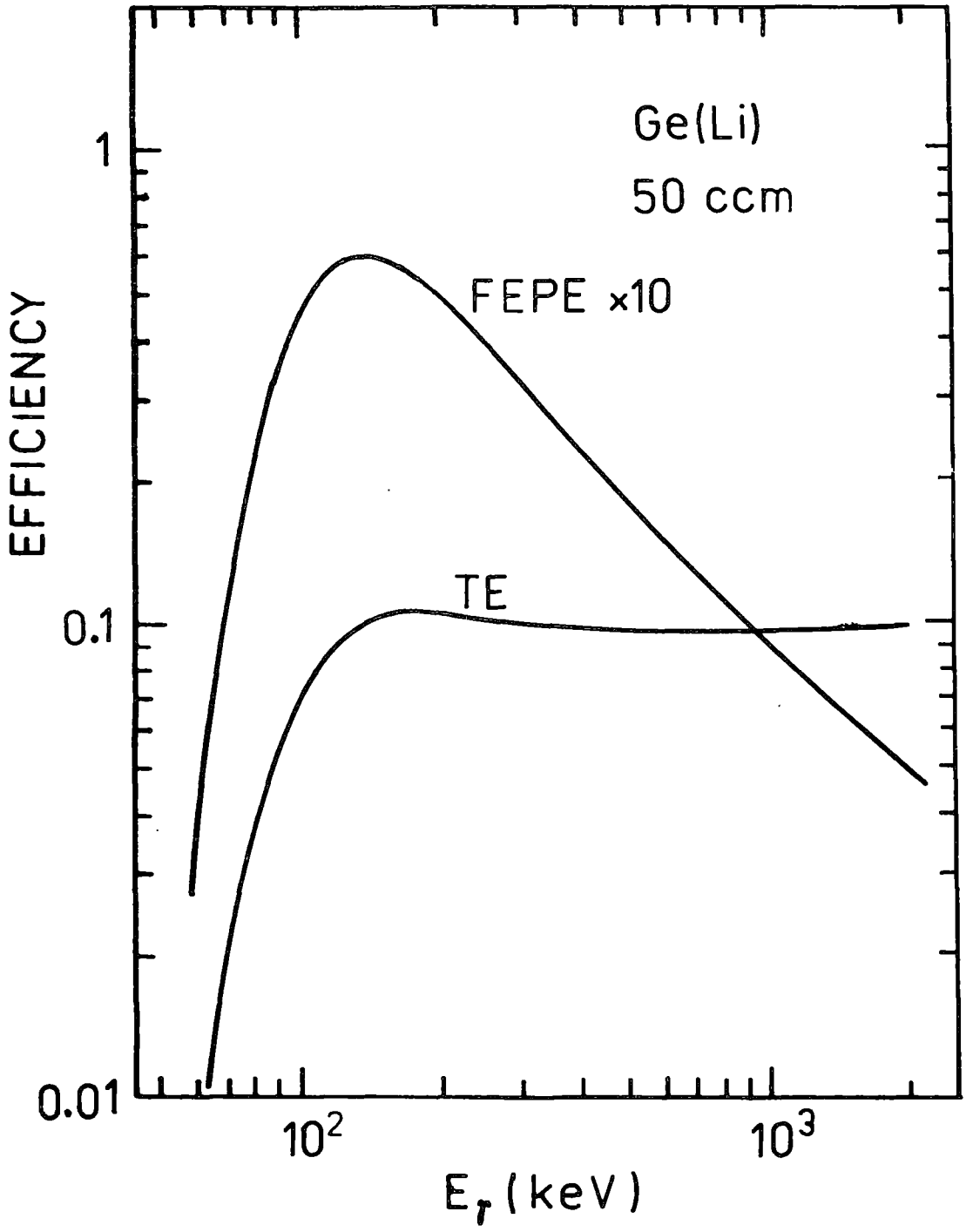

Fig. 1

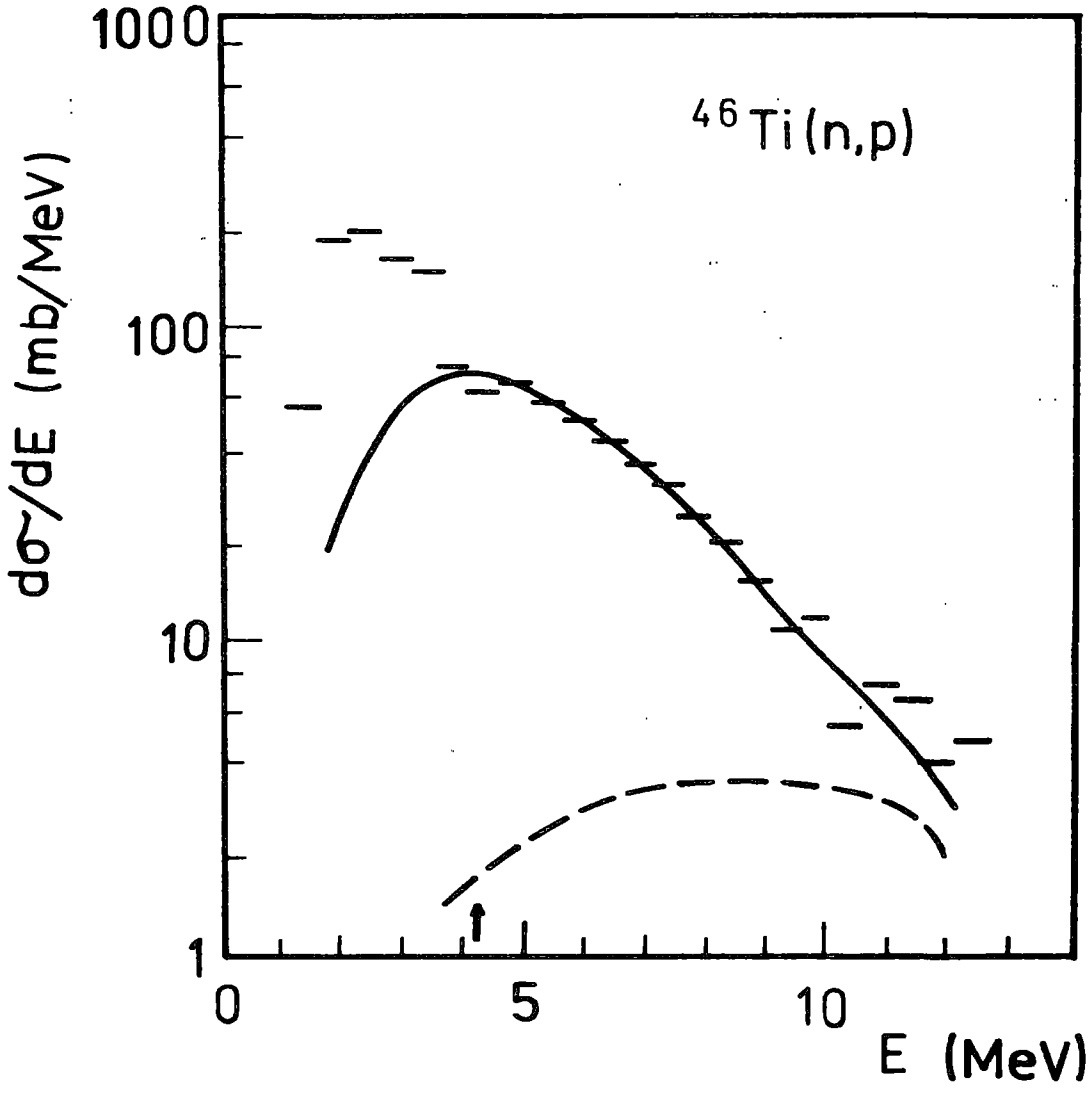

Fig. 2

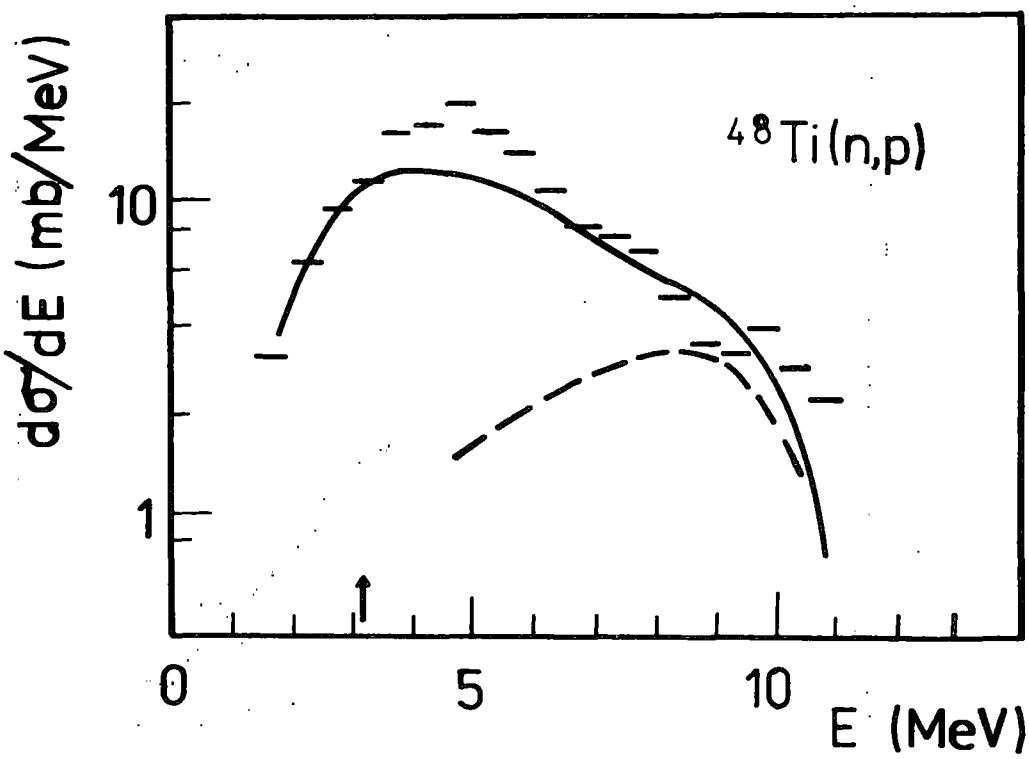

Fig. 3