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MEASUREMENTS OF THE AL-27(n,α) AND MG-24(n,p) CROSS SECTION BETWEEN 8 MEV AND 15 MEV

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Abstract: By means of deuterons impinging on a deuterium gas cell, 'monoenergetic' neutrons between 8.3 MeV and 14.7 MeV were generated with the D(d,n)He-3 reaction. Samples of aluminum and magnesium were irradiated at a distance of 9 cm from the neutron target. The samples were placed back-to-back to a U-238 fission chamber acting as a neutron fluence monitor. Empty gas cell irradiations allowed the contribution of parasitic neutrons produced in the structural material of the gas cell to be subtracted. Neutron time-of-flight spectra were recorded simultaneously with the activation procedure to determine the continuous neutron energy distribution of the D(d,np) break-up component and to correct its contribution to the activation process. The radioactivity of the samples was measured with a germanium detector calibrated against a large sodium iodide well-type detector. Below 13.6 MeV the excitation functions of both reactions were measured in steps of about 0.25 MeV. The relative uncertainties of the final data were smaller than 3 % below 11 MeV and smaller than 4 % below 12.5 MeV.

(Neutron cross section, Al-27(n,α) and Mg-24(n,p) relative to U-238(n,f), E_n = 8.3 to 14.7 MeV)

Introduction

The Al-27(n,α) reaction cross section is regarded as a fast neutron standard cross section /1/. While this cross section is well established around 14 MeV neutron energy, between 9 MeV and 12.5 MeV the available data base in sparse and requires additional confirmation. It was the aim of the present work to improve the quality of this cross section below 14 MeV. At 26 different neutron energies, between 8.3 MeV and 14.7 MeV, the Al-27(n,α) cross section was measured using the well-known fission cross section of U-238 as a reference. With the same experimental procedure the Mg-24(n,p) cross section was also determined.

Experimental Methods

Quasi-monoenergetic neutrons were produced via the D(d,n)He-3 reaction with deuterons of 5.5 MeV to 12.2 MeV, extracted from the PTB compact cyclotron CV-28. A gas target, 11 mm in diameter and 30 mm in length, was used. The gas cell was filled with deuterium gas at a pressure of 2000 hPa. The entrance window consisted of a 5.5 µm molybdenum foil and a 0.5 mm thick gold disk cooled by streaming air acted as a beam stop. Neutrons from 8.3 MeV to 14.7 MeV were so obtained. The neutron energy was determined by the time-of-flight method using a flight path of 12 m between the target and an NE213 scintillation detector 102 mm in diameter and 25 mm in length. A parallel plate fission chamber (55 mm in diameter) was mounted in the zero degrees direction at a distance of 91 mm from the middle of the gas target. The fission deposit (U-238, enriched to 99.98 %) of about 200 µg/cm^2 was on an aluminum backing, 0.2 mm thick and 20 mm in diameter. The diameter of the deposit was 10 mm. The backing was attached to the 0.3 mm thick steel front window of the fission chamber and was centered with nylon screws. The distance between the fission deposit and the electrode (brass, 0.5 mm thick) was about 10 mm. The chamber was operated with continuously flowing argon gas at atmospheric pressure at a voltage of 500 V. In a back-to-back geometry, high purity metallic foils of aluminum (99.999 %) and magnesium (99.9 %), 10 mm in diameter and 1 mm thick, were fastened in front of the fission chamber and centered to the position of the fission deposit. One aluminum and one magnesium disk were activated per irradiation. The typical duration of one irradiation run was 3 hours with a deuterium beam current of about 2 µA. For a fixed deuterium energy, each irradiation was followed by a second run with an empty gas cell to determine the contribution of secondary neutrons generated outside the deuterium gas cell. The time variation of the neutron fluence was monitored with a 38 mm x 38 mm Ø NE213 detector located at a distance of 6.5 m and turned towards the gas target at an angle of 60 degrees through a collimating system. The detector events and the integrated beam charge were recorded at periodic intervals.

Data Analysis

Radioactivity Counting

The Na-24 radioactivity of the aluminum and magnesium samples was measured with a 130 cm^3 Ge(Li) detector. The samples were counted at a distance of 16 mm from the detector face. The photopeak of the 1368.6 keV transition of Na-24 was analyzed. The efficiency curve was established with monoenergetic radioactive standard sources. Corrections were applied for the radial and axial dependence of the efficiency over the whole volume of the samples and for self-absorption within the samples. Due to the close sample-to-detector geometry, the correction for summing coincidence losses was 9.7 %.

One aluminum and one magnesium sample were also measured with a large, integral counting

NaJ(Tl) well-type detector /2,3/. The radioactivity derived agreed within 0.4 % with that obtained with the Ge(Li) detector. An appropriate normalization factor was applied to all Ge(Li) detector measurements. The measured radioactivities were converted into reaction rates per atom taking the foil masses into account and using a value of (14.9575 ± 0.0029) h for the Na-24 half-life. For the photon emission probability a value of 0.99994 ± 0.00002 was used and isotopic abundances of 1 and 0.7899 were assumed for Al-27 and Mg-24, respectively. The maximum correction in the activity build-up factor due to variations of the neutron fluence was 0.4 %.

Fission Rates

Fission fragment spectra were continuously accumulated during an irradiation. These spectra were integrated above a threshold discriminating against α-particles and noise. Below the threshold the spectra were horizontally extrapolated to zero energy. This correction was typically of the order of 1.5 %. Corrections for fission fragment losses were calculated and were between 0.85 % for the lowest and 0.30 % for the highest neutron energy.

Fission Deposit Mass Determination

The value of the fission deposit mass was determined in a separate experiment /4/. The fission chamber of this experiment was exposed in a T(d,n) neutron field at 14.8 MeV neutron energy. As in the previous experiment an aluminum disk was mounted in front of the chamber. The fission deposit mass was derived from the ratio of the reaction rates of U-238(n,f) to Al-27(n,α), and was based on the well-known cross sections of both reactions at 14.8 MeV. Irradiations were done at various distances from the solid-state Ti-T target to investigate the influence of room-return neutrons on the number of recorded fissions. Careful corrections for secondary neutrons were applied. Within the uncertainties, the three different runs agreed in their results. From a weighted average, a value of (168.2 ± 2.5) μg for the U-238 fission deposit mass was obtained.

First attempts to derive the deposit mass from α-counting in a 2π geometry resulted in a mass value which is about 8 % lower than the above quoted result. However, there are no precise corrections for self-absorption and backscattering, and the low α-emission rate of 2 Bq requires more, careful investigation. Further experiments are in progress.

Correction for Parasitic Neutrons

The reaction rates obtained from the empty gas cell irradiations were normalized to the same beam charge and were subtracted from those obtained with the filled gas cell. Thus the contribution of secondary neutrons produced in the structure material of the gas cell was eliminated. These corrections showed a pronounced increase with increasing neutron energy and were, in the energy range of this experiment, between 0.2 % and 7.0 % for Al-27(n,α) and between 0.2 % and 7.5 % for Mg-24(n,p). In the case of U-238(n,f) the high sensitivity of this reaction to low-energy neutrons resulted in corrections ranging from 1.2 % at 8.3 MeV to 23.7 % at 14.7 MeV.

Correction for Break-up Neutrons

Above 4.45 MeV incident deuteron energy, the D(d,n) reaction shows an increasing neutron yield due to break-up reactions. The broad energy distribution of these neutrons is separated by an energy gap of about 6.5 MeV from the 'mono-energetic' neutron peak (Fig. 1). Neutron energy spectra for the D + d interaction were systematically measured /5/ for deuteron energies from 5.3 MeV to 13.3 MeV (in steps of about 0.5 MeV) and for neutron emission angles between 0 degrees and 15 degrees (in steps of 2.5 degrees). These spectra were folded with the excitation functions of Al-27(n,α), Mg-24(n,p) and U-238(n,f) and for each reaction the break-up neutron contribution relative to that of the mono-energetic peak was determined. These factors are plotted in Fig. 2. Due to the fact that the neutron yield of break-up to monoenergetic peak is almost constant for emission angles smaller than 5 degrees, the zero degrees spectra were used in the analysis. The measured reaction rates were reduced by these factors to yield rates corresponding to the mono-energetic peak. For the maximum neutron energy of 14.74 MeV, these corrections were 0.102 for Al-27(n,α), 0.172 for Mg-24(n,p) and 1.122 for U-238(n,f). The uncertainty of these corrections was estimated as 5 % of the correction factor.

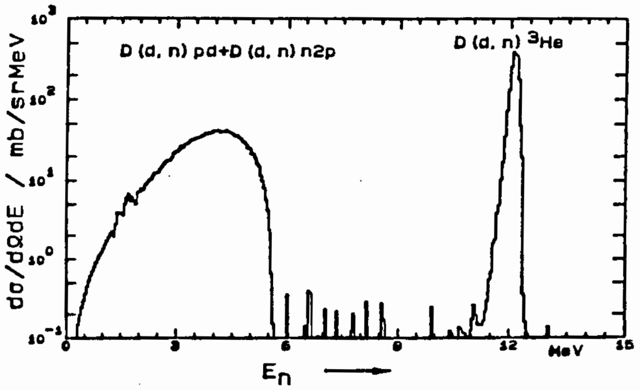

Fig. 1 Neutron energy spectrum at zero degrees for an incident deuteron energy of 8.8 MeV

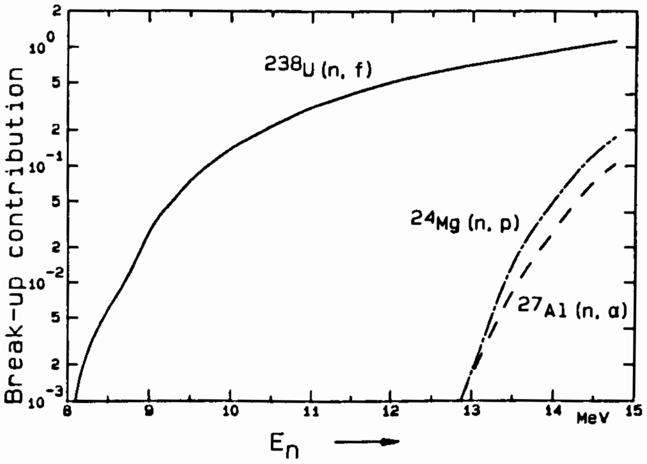

Fig. 2 Ratio of the break-up neutron contribution relative to that of the monoenergetic neutron peak for the various reactions

Neutron fluence attenuation

Due to slightly different geometrical positions of the Mg and Al disks relative to the fission deposit and due to the fluence attenuation in the various materials, corrections were applied to reduce all measured reaction rates to the same fluence. The effect of scattering within the foils, the backing and the fission chamber wall was estimated as less than 0.5 % for all neutron energies. The combined corrections were only weakly energy-dependent and were about 7.5 % for the Mg foils, always placed in front of the Al foils, and 4.2 % for the Al foils. In another experiment, these corrections were experimentally verified by replacing the Mg foil by a second Al foil. The results agreed within 0.3 %.

Results and Discussion

The corrected reaction rate ratios of Al-27(n,α) to U-238(n,f) and Mg-24(n,p) to U-238(n,f) were normalized by taking the U-238 fission cross section data from the preliminary version of ENDF/B-VI /6/. The results are plotted in Figs. 3 and 4 and are compared with recent evaluations /7,8/. The combined relative uncertainty (1 standard deviation) of the experimental data comprises the following components (only major components are listed):

Al-27(n,α):	
Counting statistics:	0.6 - 1.1 %
Efficiency•:	1.0 %
Break-up correction:	0.0 - 0.5 %

Mg-24(n,p):	
Counting statistics:	0.7 - 1.2 %
Efficiency•:	1.0 %
Break-up correction:	0.0 - 0.9 %

U-238(n,f):	
Counting statistics:	0.7 - 1.0 %
Fission deposit mass:	1.5 %
Break-up correction:	0.1 - 5.6 %
(n,f) cross section:	0.7 - 1.5 %

•Including the uncertainty of the photopeak area determination

The resulting combined uncertainty of the Al-27(n,α) and the Mg-24(n,p) cross sections ranges from 2.5 % to 6.1 % and is ≤ 3 % below 11 MeV and ≤ 4 % below 12.5 MeV neutron energy.

The Al-27(n,α) cross section data obtained in this experiment are shown in Fig. 3. Within the uncertainties they largely agree with the evaluated data of Vonach /7/, of Kornilov et al. /9/ and with the ENDF/B-V data. However, a tendency for the experimental cross sections to be higher than the evaluated data can be observed. Due to the relatively large uncertainties of the experimental data around 14 MeV resulting from the break-up correction of the U-238 fission rates, a direct comparison with the evaluated data, which are of a high accuracy in this range, is difficult. The energy resolution used in the experiment was between 150 keV and 90 keV for 8.3 MeV and 14.7 MeV, respectively. Nevertheless, a slight structure in the experimental data can be seen, the shape of which fits well with the whole body of available data (see /7/, for example) and is very similar to the structure shown by the data of Butler et al. /10/ below 12 MeV and to that of Ferguson et al. /11/ above 12 MeV.

The Mg-24(n,p) cross section data are plotted in Fig. 4. The evaluation of Vonach et al. /8/ is shown for comparison. Again, there is agreement between the experimental data and the evaluation within the uncertainties, but the same tendency as shown by Al-27(n,α) towards slightly higher experimental values compared with the evaluated data can be seen. The cross section of this reaction shows a pronounced structure. Between 8.5 MeV and 12 MeV only a single experiment /10/ can be used for comparison with the present data (see also /8/). Both experiments agree in

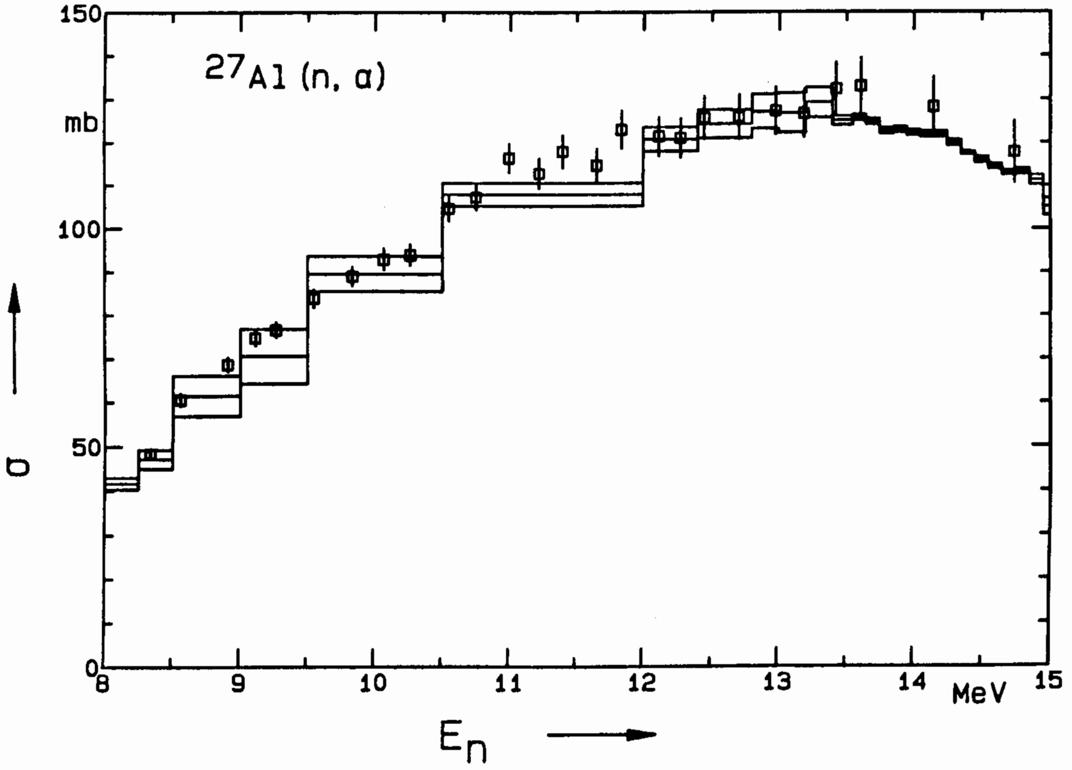

Fig. 3 Experimental data of the Al-27(n,α) cross section compared with the evaluation of Tagesen and Vonach /7/. The upper and lower line of the histogram indicates the standard deviation of the evaluated data.

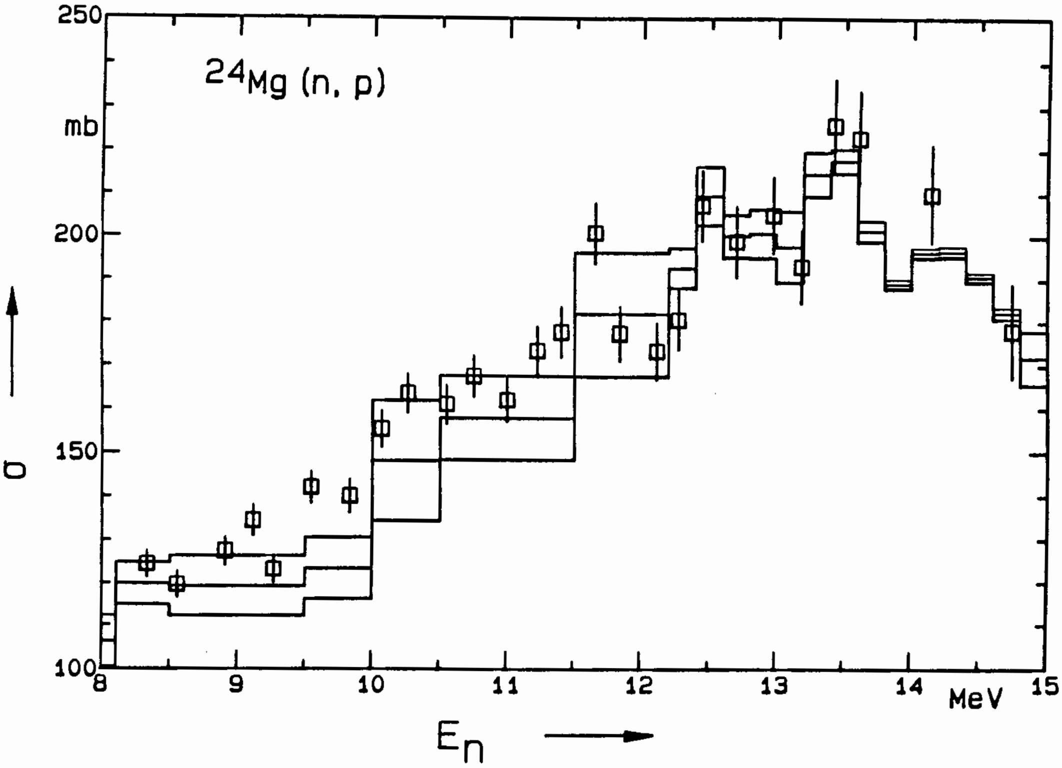

Fig. 4 Experimental data of the Mg-24(n,p) cross section compared with the evaluation of Vonach et al. /8/. Same representation as in Fig. 3.

the relative shape of the excitation function and above 12 MeV, there is similar agreement with the data of ref. /11/.

The Al-27(n,α) and Mg-24(n,p) cross section data of this work strongly depend on the validity of the fission deposit mass determination. Unlike the D(d,n) neutron field, for the T(d,n) neutron field used for the mass determination, there are unfortunately no measured neutron energy spectra available. Due to the high sensitivity of the U-238(n,f) cross section to low energy neutrons, it cannot be excluded that the measured fission rates in the mass determination experiment are disturbed by unknown low energy neutron components. Disregarding the uncertainties, an average energy-independent bias factor between experiment and evaluation /7,8/ was derived for the present data. For Al-27(n,α) and for Mg-24(n,p), the same result of a bias factor of 1.045 was obtained, while for the directly formed ratio of the Al-27(n,α) data relative to the Mg-24(n,p) data, the bias factor between experiment and evaluation disappeared. This all supports the conclusion that an inconsistency arises from the U-238 fission rates. The U-238(n,f) cross section is believed to be well established and because there is no pronounced energy-dependence of the bias factor, the break-up neutron correction factors can also be excluded as a possible error source. Finally, only the fission deposit mass determination remains to explain the suspected inconsistency of the data. To substantiate the above arguments, more experiments are needed to consolidate the value of the fission deposit mass.

Conclusions

Below 13 MeV neutron energy, the Al-27(n,α) and Mg-24(n,p) cross section data determined in this experiment constitute a valuable contribution to the existing data base. The data obtained above 13 MeV cannot compete with those from other high precision experiments and were mainly determined to give some overlap between the various experiments.

It has been demonstrated that the D(d,n) neutron field is suitable for use as a monoenergetic neutron source for cross section measurements up to 15 MeV neutron energy when data from careful investigations of the neutron break-up component are available.

To make full use of the precision of the present data and for their inclusion in future evaluations, it was found that more experiments to establish the U-238 fission deposit mass of this experiment are necessary.

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