MEASUREMENTS OF THE NEUTRON CROSS SECTION FOR Fe-54(n,alpha)Cr-51 BETWEEN 5.3 AND 14.6 MEV

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Abstract: Measurements of the neutron cross sections of Fe-54(n,α)Cr-51 and of Fe-54(n,p)Mn-54 are reported. Two experiments were performed, one at ANL between 5.3 and 9.8 MeV and one at PTB between 9.1 and 14.6 MeV. In both experiments the D(d,n)He-3 reaction served as the neutron source reaction. The neutron fluence was monitored with the Al-27(n,α)Na-24 reaction in the PTB experiment and with a U-238 fission chamber in the ANL experiment. Details of the data corrections are discussed and the results are compared with ENDF/B-VI and data from the literature.

(Neutron cross section measurements, Fe-54(n,α)/U-238(n,f) and Fe-54(n,p)/U-238(n,f), E_n = 5.3 to 9.8 MeV; Fe-54(n,α)/Al-27(n,α) and Fe-54(n,p)/Al-27(n,α), E_n = 9.1 to 14.6 MeV)

Introduction

The Fe-54(n,α)Cr-51 reaction contributes to the helium accumulation in steel, and this is of some technical importance in intense, fast neutron fields. The data base of this cross section is sparse (in the threshold region, for example) and also contradictory, and there is a complete absence of data between 10 and 12.5 MeV. To improve our knowledge of this cross section, measurements have been made between 5.3 and 14.6 MeV.

The measured cross section ratios were normalized with cross section values taken from ENDF/B-VI for U-238(n,f) and from a recent evaluation [1] for Al-27(n,α). The two monitor reactions are related by a recent Al-27(n,α) to U-238(n,f) experiment [2] which is part of the Al-27(n,α) evaluation [1].

Experimental Methods

ANL Experiment

At the ANL Fast Neutron Generator Facility, a deuterium gas target was used for the neutron production. The water-cooled target, 2.54 cm in diameter and 2 cm long, was pressurized at 0.2 MPa. With deuterons betwen 2.5 MeV and 7 MeV, neutrons from 5.3 to 9.8 MeV were produced. A low-mass fission chamber was placed at zero degrees at a distance of 3.2 cm from the target. An enriched U-238 deposit (< 6 ppm of isotopes other than U-238) of 386 μg/cm² and 2.54 cm in diameter was employed for the fluence measurements. A detailed specification of this deposit, sample no. U-238-60, is given elsewhere [3]. Natural iron samples of 98.5 % purity, 2.54 cm in diameter and 2.9 mm thick, were irradiated back-to-back to the fission deposit in front of the fission chamber. Separate runs with an empty gas cell were conducted to determine the neutron background from the beam stop (1.9 mm of Au) and the walls of the cell. The neutron energy resolution (FWHM) of this experiment was between 0.16 and 0.27 MeV. The neutron energy scale was calibrated with protons by observing the thresholds for the Li-7(p,n) and B-11(p,n) reactions.

PTB Experiment

Deuterons between 6.3 MeV and 12.0 MeV, extracted from the PTB compact cyclotron CV-28, interacted with a deuterium gas target 1.1 cm in diameter and 3 cm long. Neutrons between 9.1 and 14.6 MeV were produced via the D(d,n)He-3 reaction. The target cooled by streaming air worked with a gas pressure of 0.2 MPa. Disks of high-purity metallic foils 1 cm in diameter, of alumimum (99.999 %) and iron (99.99+ %) were irradiated at a distance of 3.9 cm from the target. Each iron foil (2 mm thick) was sandwiched between two aluminum foils 1 mm thick. The 'gas-out' corrections ranged from 0 to 3.6 % for Fe-54(n,α), from 1.5 % to 21 % for Fe-54(n,p) and from 0.2 % to 9.5 % for Al-27(n,α). Before each irradiation the neutron energy was measured by the time-of-flight method, operating the cyclotron in a pulsed mode (about 1 ns pulse width and 1 MHz repetition frequency). The flight path between the target and an NE213 neutron detector 10.2 cm in diameter and 2.54 cm thick, was 12 m. With the Monte Carlo code SINENA [4] simulating the neutron production in the gas target, the neutron energy measured and its distribution was transformed to that of the close sample-to-target geometry. The energy resolution was between 174 keV and 204 keV (FWHM).

Data Analysis

Radioactivity counting

The Cr-51 and Mn-54 radioactivity of the iron samples was measured with various Ge(Li) detectors. In the ANL experiment three different detectors were involved allowing fairly long counting times per sample. The sample-to-detector distances varied from 1 to 2 cm. At PTB, a single detector was used with a sample distance of 1.6 cm. The nominal efficiencies were corrected for sample size effects (radial and axial dependence) and for self-absorption within the samples. A PTB standard source of Cr-51 was used to intercompare the various detectors. In addition, a few iron samples were measured at ANL as well as at PTB. For the measured photopeaks of 320.1 keV (Cr-51) and 834.8 keV (Mn-54) a fair consistency between the various independently calibrated detectors was found which confirmed the calibration uncertainties of the ANL detectors (2 % for Cr-51 and 1.5 % for Mn-54) and of the PTB detector

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(1.5 %). The Na-24 activity of the aluminum foils of the PTB experiment was measured at a sample-to-detector distance of 5.3 cm, reducing the correction for summing coincidence losses from 9.7 % to 2.4 %. Other details are given in Ref. [2]. Recommended decay parameters [5] for the half-life ($T_{1/2}$) and the photon emission probability (h_γ) were used:

Cr-51 $T_{1/2}$ = 27.71(3) d h_γ = 0.0985(9)
Mn-54 $T_{1/2}$ = 315.5(5) d h_γ = 0.99975(3)

A value of (5.8 ± 0.1) % was adopted for the isotopic abudance of Fe-54 [6].

Influence of scattered neutrons

Neutrons scattered from the wall of the gas target, from support structures, the sample holder, the fission chamber material and from neighboured samples and also scattered within the samples, increase the reaction rates of the nuclear reactions investigated. To take this into account, Monte Carlo calculations were done which determined the individual contributions to each reaction. The code described in Ref. [7] simulates elastic and inelastic scattering processes. Collisions of a second and higher order were neglected, an approximation which is valid as long as the neutron transmission through the scatterer is large.

In the ANL experiment these corrections were between 12.7 % (5.3 MeV neutrons) and 8.9 % (9.8 MeV neutrons) for U-238(n,f), between 7.6 % and 5.6 % for Fe-54(n,p) and between 5.9 % and 4.1 % for Fe-54(n,α). In the PTB experiment, with a different geometry the corrections were between 3.8 % (9.1 MeV neutrons) and 3.3 % (14.6 MeV neutrons) for Al-27(n,α), between 5.7 % and 5.4 % for Fe-54(n,p) and between 4.1 % and 3.5 % for Fe-54(n,α). In ratio measurements only the difference between these values remains as a final correction factor.

Correction for breakup neutrons

A second effect which increases the reaction rates is due to breakup neutrons of the D(d,np) reaction. At the maximum neutron energy of 9.82 MeV used in the ANL experiment, these corrections were 10.5 % for U-238(n,f), 1.8 % for Fe-54(n,p) and negligible for Fe-54(n,α). In the PTB experiment the corrections at 14.64 MeV were 9.7 % for Al-27(n,α) and 21.9 % for Fe-54(n,α). For Fe-54(n,p) the correction is extreme, with a ratio of the contribution to the radioactivity of 2.628:1 for the breakup neutrons compared with the 14.64 MeV neutrons. These corrections were calculated on the basis of detailed measurements of breakup neutron spectra. The measurements covered the deuteron energy range between 5.3 and 13.3 MeV and were done for neutron emission angles up to 15 degrees [8]. Details and examples are given in the reference. The uncertainty of the corrections was estimated at 3 % of the amount of the correction.

Final uncertainties

Further uncertainty components which have been included were the time variation of the empty gas cell correction (0.5 - 2.5 %), the non-uniform distribution of the activity in the samples (0.5 %), impurities of the sample material (0.0 - 0.5 %), fission fragment losses (1 %) and extrapolation to zero bias (1.1 %), the deposit mass (0.7 %), geometry factors

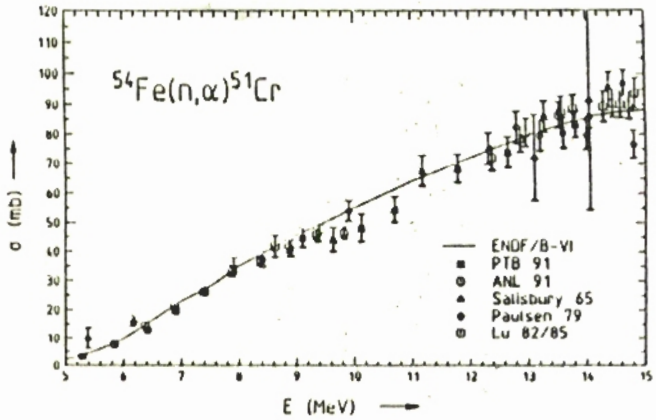

Fig. 1. Experimental data from the present work (ANL91 and PTB91) for the Fe-54(n,α) reaction

between the different sample positions (1.4 %) and the influence of the energy scale uncertainty. The last component is negligible for the relatively flat Fe-54(n,p) cross section but amounts to between 9.0 % and 0.1 % for the Fe-54(n,α) reaction in the energy range from 5.3 to 9.8 MeV. The uncertainties of the normalization cross sections were between 0.7 and 0.9 % for U-238(n,f) and between 0.5 and 3.2 % for Al-27(n,α).

The final uncertainties (one standard deviation) were obtained by combining the individual components quadratically. The range of the results was: 4.1 % - 13.3 % (0.8 % - 9.1 %) for Fe-54(n,α) and 3.2 % - 4.7 % (0.2 % - 3.4 %) for Fe-54(n,p) in the ANL experiment and 4.2 - 9.7 % (2.5 - 8.6 %) for Fe-54(n,α) and 3.8 - 8.6 % (0.8 - 1.4 %) for Fe-54(n,p) in the PTB experiment. The values of the counting statistics are separately listed in brackets. The large uncertainties of the Fe-54(n,p) data of the PTB experiment at high neutron energies reflect the amount of the breakup correction, which strongly increases with the energy.

Results and Conclusions

The data from the present work for the Fe-54(n,α) reaction between 5.3 and 14.6 MeV are plotted in Fig. 1. They are compared with the ENDF/B-VI evaluation and data from the few other experiments which have covered a similarly large energy range. The references were taken from the CINDA index [9]. A fair agreement between the ANL and the PTB experiment was found in the overlap region. With the exception of the radioactivity counting, in each of the two experiments slightly different experimental techniques and data analysis methods were used, which indicates a high degree of reliability for both experimental procedures. Below 9 MeV, the present data are in fair agreement with the ENDF/B-VI evaluation. Around 10 MeV, our data indicate a shoulder in the excitation function and are about 15 % lower compared with ENDF/B-VI. At high neutron energies, our data show a larger slope than the ENDF/B-VI evaluation. This trend is in agreement with another data set (Lu82/85). All other experiments were undertaken with 14 MeV neutrons. This is shown in Fig. 2 where all post-1975 experiments are plotted and compared with our data and with ENDF/B-VI. Above 14.3 MeV, our data are about 10 % higher than

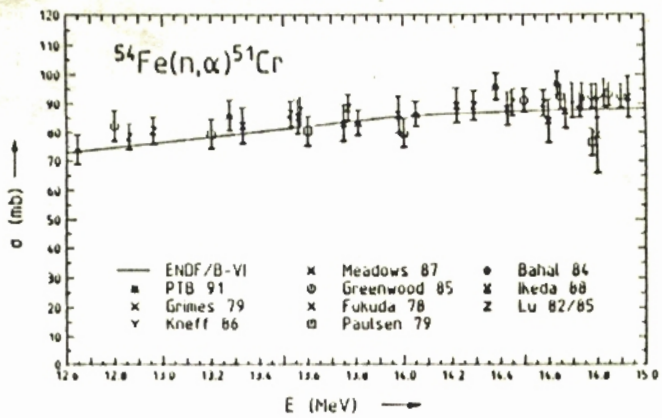

Fig. 2. The Fe-54(n,α) reaction between 12.5 and 15 MeV

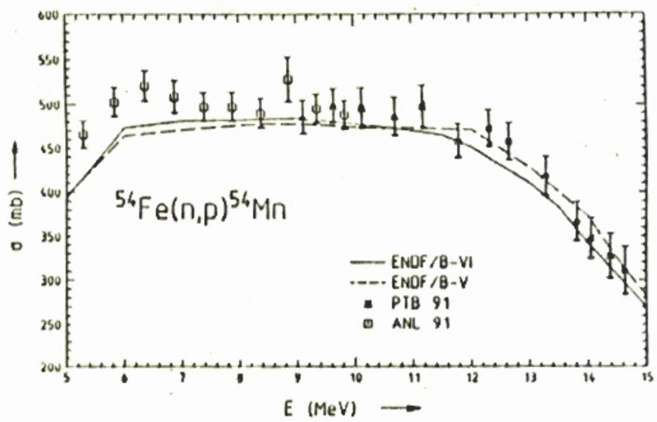

Fig. 3. Experimental data from the present work for the Fe-54(n,p) reaction

ENDF/B-VI and also higher than most of the data from other experiments.

In Fig. 3 our data on the Fe-54(n,p) reaction are plotted. For this reaction too, the agreement between the ANL and PTB experiment in the overlap region is very satisfactory. Above 7 MeV most of our data agree within the uncertainties with the ENDF/B-VI evaluation. Nevertheless, our data show a general tendency to be about 3 - 4 % higher than ENDF/B-VI. At the low energies in Fig. 3 this deviation amounts to 10 %. At high neutron energies substantial correction factors for breakup neutrons were applied to our data. A useful test of the validity of these corrections is a comparison with the 14 MeV data obtained with the T(d,n)He-4 neutron production reaction which is free of such corrections. There is usually a large amount of data for around 14 MeV (see Fig. 2, for example). A disadvantage is that these data very often show a large spread in their values and there is no guarantee that the average of these data represents a 'true' result. Two recent evalutions of 14 MeV data [10, 11] were used for a comparison. Both were performed at 14.70 MeV. Our 14.64 MeV data were extrapolated to this energy by using the individual slopes of the ENDF/B-VI evaluation for each reaction. The result is summarized in Table 1. The PTB experiment also comprised a measurement of the Fe-56(n,p) reaction which is not shown in the present work. The corresponding 14.64 MeV data point is included in the comparison in Table 1.

Within the uncertainties there is agreement between our data and the evaluations. For the Fe-54(n,α) reaction the agreement is at least within two standard deviations. However, our data on Fe-54(n,α) and Fe-54(n,p) are about 10 % and 8 % higher than the evaluated data. There is no clear correlation of these deviations with the magnitude of the breakup corrections. The net correction for Fe-54(n,α) is 12.2 %, the difference between a 21.9 % correction of this

Table 1. Comparison with evaluated cross sections (in mb) at 14.70 MeV

Reaction	Present work*	Evaluation	Ref.
Fe-54(n,α)	97.2 ± 4.0	87.9 ± 2.4	[10]
Fe-54(n,p)	306.2 ± 26.7	284.4 ± 5.7	[10]
Fe-56(n,p)	109.0 ± 3.1	108.4 ± 0.5	[11]

*Extrapolated from 14.64 MeV to 14.70 MeV

reaction and a 9.7 % correction of the Al-27(n,α) monitor reaction. The Fe-56(n,p) reaction has a very similar net correction of 11.9 %. But for the Fe-54(n,p) reaction the breakup correction is an order of magnitude larger. Only 27.5 % of the measured reaction rate is from the 14.64 MeV neutrons.

The agreement shown in Table 1 is very encouraging. It indicates that also very large breakup corrections can be performed at a high level of confidence. Data obtained with the D(d,n)He-3 reaction are rarely superior to those measured with the T(d,n)He-4 reaction. This is due to the increasing uncertainties resulting from the breakup corrections. But in the intermediate region between 10 and 13 MeV neutron energy, the quality of data measured with the D(d,n)He-3 reaction does not seem to be seriously handicapped by corrections for breakup neutrons.

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