

## SYSTEMATIC MEASUREMENT OF NEUTRON ACTIVATION CROSS SECTION AT ENERGY RANGE FROM 13.3 TO 15.0 MEV USING FNS FACILITY

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Abstract: A hundred and forty neutron activation cross sections at energy range from 13.3 to 15.0 Mev have been measured systematically at FNS facility for various structural materials of fusion reactor. The large body of data measured on the unified experimental condition allow the significant improvement of data accuracy required in many applications. Several dosimetry reaction cross sections based on the present data gave consistent results in the blanket integral experiment. It was proved that the present measurement could provide more reliable data base for the formulation of systematic trend for principal reactions in comparison with data previously reported in literatures.

(Activation cross section, Structural material, Fusion reactor, (n,p), (n,n'p), (n, $\alpha$ ), (n,2n), Blanket integral experiment, Systematic trend, Isomeric cross section ratio)

Introduction

A program of activation cross section measurement at a neutron energy range from 13.3 to 15.0 MeV for fusion reactor structural materials has been conducted using FNS facility/1/. Up to now, 140 different reaction cross sections have been measured on an unified experimental conditions. The 110 data of all were already reported 2/. Primary objectives of the program were to provide

new experimental data with adequate accuracy needed in the fusion applications such as the dosimetry study, the induced activity analysis and so on, and then to obtain the more accurate data base for formulating the systematic trend of the principal reactions in fusion neutron fields. First, the experimental procedure is briefly given. The data are discussed mainly in the framework of the systematic trend for the cross section at around 14 MeV neutron energy.

Table 1 Reaction types and Target isotopes

(n,p):	$^{24}\text{Mg}$ , $^{25}\text{Mg}$ , $^{27}\text{Al}$ , $^{28}\text{Si}$ , $^{29}\text{Si}$ , $^{41}\text{K}$ , $^{42}\text{Ca}$ , $^{43}\text{Ca}$ , $^{44}\text{Ca}$ , $^{46}\text{Ti}$ , $^{47}\text{Ti}$ , $^{48}\text{Ti}$ , $^{50}\text{Ti}$ , $^{51}\text{V}$ , $^{52}\text{Cr}$ , $^{54}\text{Fe}$ , $^{56}\text{Fe}$ , $^{59}\text{Co}$ , $^{58}\text{Ni}$ , $^{60}\text{Ni}$ , $^{61}\text{Ni}$ , $^{65}\text{Cu}$ , $^{67}\text{Zn}$ , $^{75}\text{As}$ , $^{85}\text{Rb(m)}$ , $^{87}\text{Rb}$ , $^{90}\text{Zr(m)}$ , $^{91}\text{Zr(m)}$ , $^{92}\text{Zr}$ , $^{94}\text{Zr}$ , $^{92}\text{Mo(m)}$ , $^{94}\text{Mo(m)}$ , $^{95}\text{Mo(m,g)}$ , $^{96}\text{Mo}$ , $^{97}\text{Mo(m,g)}$ , $^{98}\text{Mo(m)}$ , $^{114}\text{Sn(m)}$ , $^{116}\text{Sn(m)}$ , $^{117}\text{Sn(m,g)}$ , $^{135}\text{Ba(m)}$ , $^{136}\text{Ba}$ , $^{138}\text{Ba}$ , $^{184}\text{W}$
(n,n'p):	$^{25}\text{Mg}$ , $^{29}\text{Si}$ , $^{44}\text{Ca}$ , $^{47}\text{Ti}$ , $^{48}\text{Ti}$ , $^{49}\text{Ti}$ , $^{57}\text{Fe}$ , $^{58}\text{Ni}$ , $^{62}\text{Ni}$ , $^{68}\text{Zn}$ , $^{91}\text{Zr(m)}$ , $^{92}\text{Zr(m)}$ , $^{94}\text{Zr}$ , $^{96}\text{Mo(m,g)}$ , $^{97}\text{Mo}$ , $^{98}\text{Mo(m,g)}$ , $^{117}\text{Sn(m)}$ , $^{137}\text{Ba}$
(n, $\alpha$ ):	$^{27}\text{Al}$ , $^{30}\text{Si}$ , $^{41}\text{K}$ , $^{44}\text{Ca}$ , $^{45}\text{Sc}$ , $^{50}\text{Ti}$ , $^{51}\text{V}$ , $^{54}\text{Fe}$ , $^{59}\text{Co}$ , $^{68}\text{Zn}$ , $^{75}\text{As}$ , $^{85}\text{Rb}$ , $^{90}\text{Zr(m)}$ , $^{94}\text{Zr}$ , $^{96}\text{Zr}$ , $^{93}\text{Nb(m)}$ , $^{92}\text{Mo(m,g)}$ , $^{98}\text{Mo}$ , $^{100}\text{Mo}$ , $^{120}\text{Sn(m,g)}$ , $^{138}\text{Ba}$ , $^{186}\text{W}$
(n,2n):	$^{19}\text{F}$ , $^{23}\text{Na}$ , $^{35}\text{Cl(m)}$ , $^{39}\text{K}$ , $^{40}\text{Ca}$ , $^{45}\text{Sc(m,g)}$ , $^{46}\text{Ti}$ , $^{50}\text{Cr}$ , $^{52}\text{Cr}$ , $^{59}\text{Co}$ , $^{58}\text{Ni}$ , $^{63}\text{Cu}$ , $^{65}\text{Cu}$ , $^{64}\text{Zn}$ , $^{66}\text{Zn}$ , $^{75}\text{As}$ , $^{85}\text{Rb(m,g)}$ , $^{87}\text{Rb}$ , $^{86}\text{Sr(m,g)}$ , $^{88}\text{Sr(m)}$ , $^{89}\text{Y}$ , $^{90}\text{Zr(m,g)}$ , $^{96}\text{Zr}$ , $^{93}\text{Nb(m)}$ , $^{92}\text{Mo(m)}$ , $^{94}\text{Mo(m)}$ , $^{100}\text{Mo}$ , $^{107}\text{Ag(m)}$ , $^{112}\text{Sn}$ , $^{114}\text{Sn}$ , $^{118}\text{Sn(m)}$ , $^{124}\text{Sn(m)}$ , $^{134}\text{Ba(m)}$ , $^{136}\text{Ba(m)}$ , $^{138}\text{Ba(m)}$ , $^{181}\text{Ta(m)}$ , $^{197}\text{Au(m,g)}$ , $^{204}\text{Pb}$ , $^{232}\text{Th}$ , $^{238}\text{U}$
Others:	$^{115}\text{In(n,n')}^{115\text{m}}\text{In}$ , $^{232}\text{Th(n,f)}$ , $^{238}\text{U(n,f)}$

\* The notation of (m) indicates that the reaction produces only a metastable state.

\* The notation of (m,g) indicates that both cross sections are available for the reactions which produce the ground and metastable states.

\* The reaction of (n,n'p) includes (n,n'p), (n,d) and (n,pn).

## Experiments

The target nuclei and reaction types measured at FNS are listed in Table 1. All of activities produced in the reactions have their half-lives ranging from 1 min. to several years. The major reactions were (n,p), (n,n'p) (n, $\alpha$ ), and (n,2n) which play important roles on the D-T fusion applications. The D-T neutrons were generated via  $^3\text{T}(d,n)^4\text{He}$  by bombarding the tritium target with deuterium beam using the FNS facility. By positioning multiple samples to be irradiated around the target at angles from  $0^\circ$  to  $165^\circ$ , the incident neutron energy ranged from 13.3 to 15.0 MeV, so that the exciting function was obtained in that energy region. In general, enriched isotopes were used as the samples which had weight of about 40 mg. The neutron flux at the sample was monitored by the reactions of  $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ ,  $^{27}\text{Al}(n,p)^{27}\text{Mg}$ , and  $^{93}\text{Nb}(n,2n)^{92\text{m}}\text{Nb}$ . In order to make the measurement efficient, five Ge detectors were employed simultaneously. The detail description of the experimental procedure was given in Ref. 2.

The neutron spectra at the sample position was calculated by the Monte Carlo code MORSE-DD/3/ with a precise model of the D-T target assembly. The effective reaction energy of the incident neutron was determined by using the calculated spectra and cross section curves determined primarily using neutron energies obtained from the reaction kinematics. The estimated error of the energy was less than 100 keV.

## Results

All cross sections were obtained relative to the cross section values of  $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$  taken from the ENDF/B-V dosimetry file/4/. In order to validate all data as absolute values, an absolute measurement on this  $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$  reaction cross section at two energies of 13.3 and 15.0 MeV was carried out by means of the associated  $\alpha$ -particle counting method/5/. The cross sections of  $112.7 \pm 3.5$  and  $129.9 \pm 4.0$  mb at respective energies were obtained and they were in good agreements with data of the ENDF/B-V. This results ensured the validation of the data obtained relative to ENDF/B-V data as absolute values. The experimental errors of principal reactions used in the dosimetry study and those in the major structural material elements were within  $\pm 5\%$ .

## Discussion

### Fusion blanket integral experiment

The spectral index plays an important role on the experimental analysis for fusion blanket integral experiment/6/. Many dosimetry reactions were employed as the indices to characterize neutron fields. However, it has been pointed out that some cross sections at 14 MeV region were inadequate, resulting in severe inconsistency in the reaction rate values. Several cross sections at 14 MeV region were replaced with present cross section values measured. As a results, the inconsistency was eliminated. A detail discussion about the experimental analysis on the reaction rate is given in Ref. 7.

### Systematic trend for reaction cross section

A number of works have been carried out to formulate systematic trends for the cross sections of (n,p), (n,n'p), (n, $\alpha$ ), and (n,2n) reactions at the 14 MeV energy region/8-11/. For the (n, charged particle) reaction, it have been proved

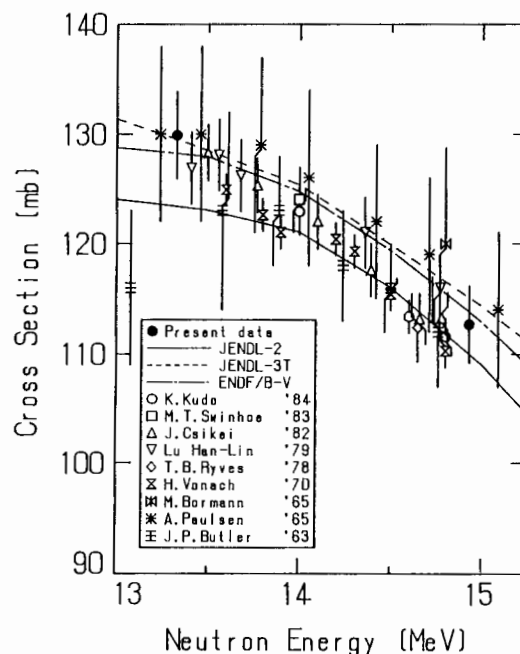


Fig. 1 Cross section data of  $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$  around 14 MeV energy region.

that the trend could be expressed as a simple function of a parameter of  $(N-Z)/A$ , where  $N$ ,  $Z$ , and  $A$  were neutron, proton, and mass numbers of the target nuclei, respectively.

### a) (n,p) reaction

For the (n,p) reaction, a large number of data at 14 MeV have been available to for formulation of the systematic trend. In Fig. 2, the present data at 14.9 MeV are plotted along with the previous data as a function of  $(N-Z)/A$ . It is evident that there is a general tendency in the data; the cross section decreases as the  $(N-Z)/A$  increases. Using present data, the systematics

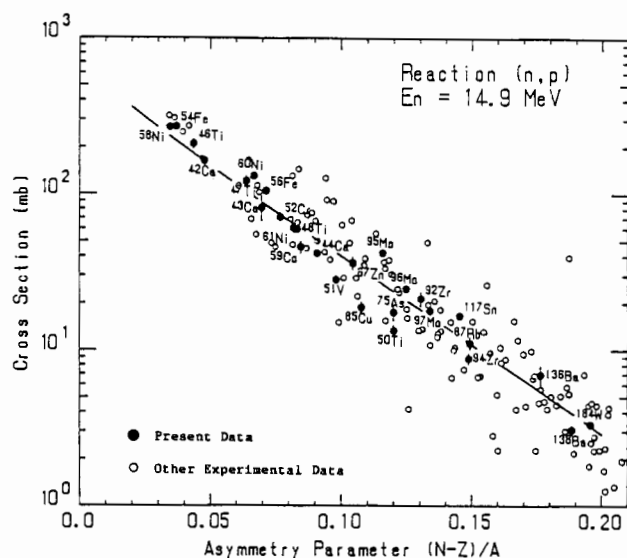


Fig. 2 Systematics of (n,p) reaction cross sections at 14.9 MeV.

was expressed by a simple formula given by,

$$\sigma_{(n,p)} = 36.18 \cdot (A^{1/3} + 1)^2 \cdot \exp(-31.468 \cdot S), \quad (1)$$

$$S = (N-Z)/A.$$

Significant is that our data lie along the line derived from eq. (1), whereas data previously reported in literatures show a rather large divergency. This result indicates that systematic cross section measurement at FNS certainly makes the analysis of such a trend very precise.

#### b) (n,n'p) reaction

In general, the (n,n'p) reaction has a steep excitation function at 14 MeV due to high threshold energy. According to the increase of energy from 13 to 15 MeV, cross section values increase rapidly to be one order of magnitude higher. In Fig. 3, the cross sections measured are plotted as a function of (N-Z)/A in the same manner of (n,p). The upper and lower curves were drawn to fit the data at 14.9 and 13.3 MeV, respectively. The data at 13.3 < E<sub>n</sub> < 14.9 MeV failed between the curves. From this result, it was suggested that a great care must be taken into the formulation of the systematic trend for (n,n'p) reaction because of high energy dependency on the cross section value. The present data covering wider energy range clearly proved importance of the energy dependency. The systematics for this reaction cross section at 14.9 MeV was expressed by,

$$\sigma_{(n,n'p)} = 680.0 \cdot (A^{1/3} + 1)^2 \cdot \exp(X), \quad (2)$$

$$X = -105.05 \cdot S + 370.59 \cdot S^2 - 471.47 \cdot S^3.$$

#### c) (n,α) reaction

Figure 4 shows the plot of the (n,α) cross sections as a function of (N-Z)/A. The data indicated by the open circle were taken from Ref. 12 which compiled available data previously reported in literatures. It is evident that the systematic trend for (n,α) is very similar to that for (n,p) reaction. The systematics is given by,

$$\sigma_{(n,\alpha)} = 24.20 \cdot (A^{1/3} + 1)^2 \cdot \exp(-34.546 \cdot S). \quad (3)$$

#### d) (n,2n) reaction

The systematics for the (n,2n) reaction cross section is shown in Fig. 5 where the data are plotted also as a function of (N-Z)/A. Contrary to the (n,charged particle) reactions, the (n,2n) gives a profile where the cross section for lighter mass region (Z < 30) increases rapidly and it increases as the mass number increases. For the data of heavy mass nuclei (A > 100), it becomes almost constant. This figure indicated that almost all data are encompassable within a narrow band along a curve which was formulated as

$$\ln \sigma_{(n,2n)} = 7.402 \cdot (1 - 1.434 \cdot \exp(-26.566 \cdot S)). \quad (4)$$

#### e) Isomeric cross section ratio

It is worthwhile to find out a systematic correlation between the isomeric cross section ratio  $[\sigma_m / (\sigma_m + \sigma_g)]$  and the spin of isomeric state J<sup>m</sup> to estimate unknown cross section values. Some works have been performed to formulate the correlation/13 - 15/ and showed that data for the (n,p), (n,α), (n,2n) and (n,t) reactions followed a parabolic trend for the ratio as a function of J<sup>m</sup>. In Fig. 6, the isomeric cross section ratios based on present experimental data are plotted against the J<sup>m</sup>. When the σ<sub>g</sub> was not available, the value

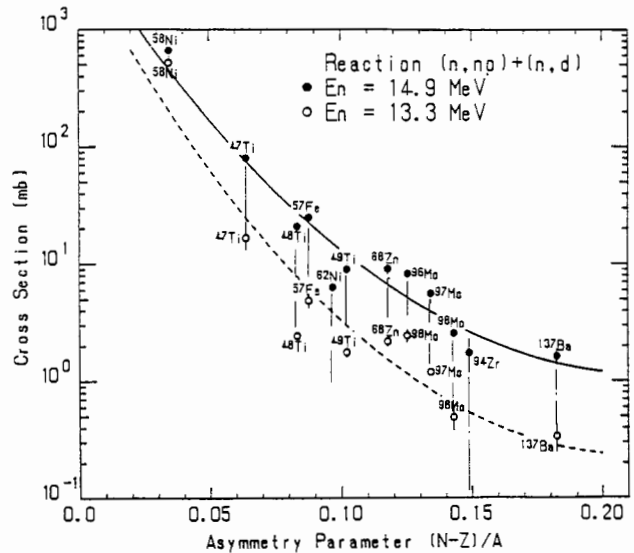
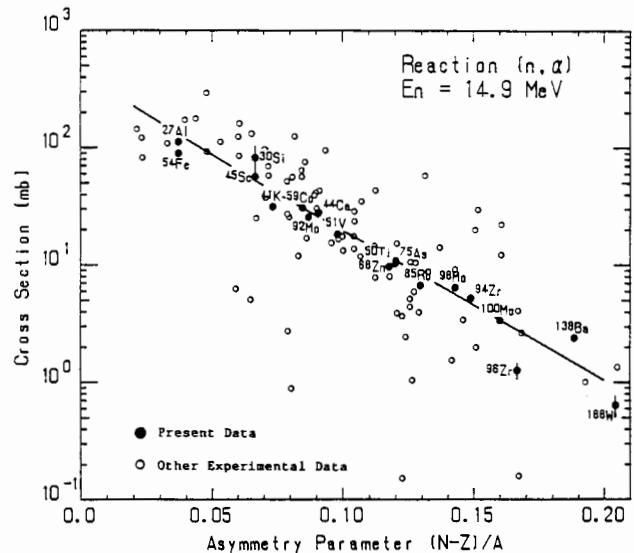


Fig. 3 Systematics of (n,n'p) reaction cross sections at 13.3 and 14.9 MeV.



was estimated from the systematic curves determined using present data. It is obviously seen that the ratios via  $J^m$  present more clearly a parabolic profile than those suggested previously. Uray/14/ suggested that the preferable spin state was 4 to 7 in the isomeric ratio. However, it should be noted that the present result indicated that the spin state of 3 to 6 is dominant in the ratio.

### Conclusion

The Activation cross sections at the neutron energy range from 13.3 to 15.0 MeV have been measured systematically by using FNS for 140 reactions induced in the structural materials. The most important result reported here is that the data based on the present systematic measurements serve to improve the consistency in the nuclear data used in the D-T fusion dosimetry as well as other applications associated with nuclear interactions. In particular, the present self-consistent data gave the reliable data base in the formulation of the systematics for the reaction cross sections around 14 MeV. Consequently, it could be concluded that the selection of reliable data for this purpose was essential.

The present experimental program is being continued extensively to obtain data for other cross sections not measured, especially for the reactions products of which have half-lives shorter than 1 min and longer than years.

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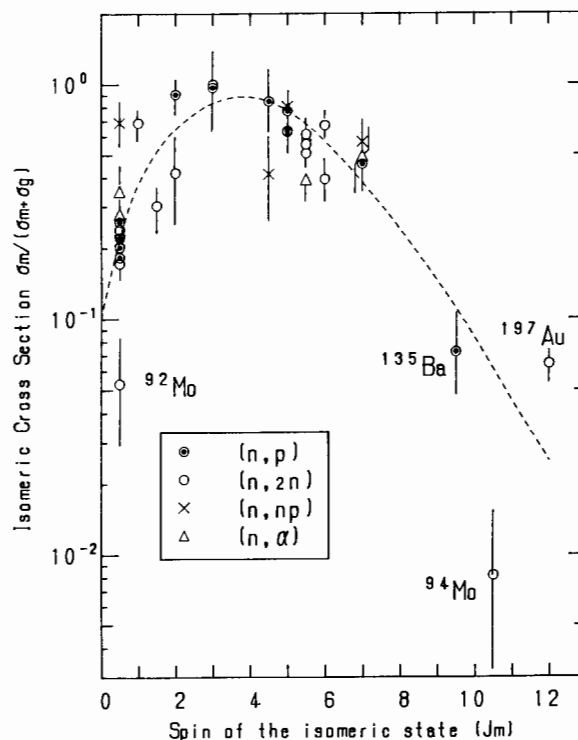


Fig. 6 Isomeric cross section ratios in (n,p), (n,n'p), (n,α) and (n,2n) at 14.9 MeV as a function of the spin of the isomeric state.