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Cross Section Measurement and Evaluation for ^{58}Ni(n,p), ^{60}Ni(n,p), ^{62}Ni(n,α) and ^{54}Fe(n,p) Reactions

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Activation technique has been used to measure the cross section for ^{58}Ni(n,p), ^{60}Ni(n,p) and ^{62}Ni(n,α) reactions in the neutron energy range 13.6-17.8 MeV. The covariance matrixes for measurement error are calculated. The range of measurement error is 3%-7%. Evaluations for ^{58}Ni(n,p), ^{60}Ni(n,p), ^{62}Ni(n,α) and ^{54}Fe(n,p) cross section are made.

1. INTRODUCTION

Both Ni and Fe are important component elements of alloy and are widely used as reactor materials and radiation protection shielding materials. It is therefore very important to measure the cross sections of their (n,p) and (n,α) reactions accurately for determining the radiation resistant ability of metals. In compliance with the requirement suggested by Consultative Group of IAEA in 1986 (Gaussing, Germany) for the data of D-T fission reaction, we measured some data of Ni which have not yet satisfied the demand in design of fission reactors. For the same purpose we also evaluated the cross sections for ^{58}Ni(n,p), ^{60}Ni(n,p), ^{62}Ni(n,α) and ^{54}Fe(n,p) reaction.

In Figs. 1-4 we give the measurement data of different authors. It can be seen that in the energy range of 15-20 MeV the data of ^{58}Ni(n,p) reaction show discrepancy, while there are few measurement data for ^{60}Ni(n,p) and ^{62}Ni(n,α) reactions. To clarify the discrepancy and to obtain supplemental data it is necessary to measure cross sections for these reactions. For ^{54}Fe(n,p) reaction the early data also showed discrepancy and some new data were given recently, hence a re-evaluation of its excitation curve is necessary.

2. MEASUREMENT

Cross sections for ^{58}Ni(n,p)$^{58m+g}$Co, ^{60}Ni(n,p)^{60}Co and ^{62}Ni(n,α)^{59}Fe reactions were measured with activation technique and the cross section of ^{27}Al(n,α)^{24}Na reaction[1] was used as relative

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Table 1
Half-life, γ-ray energy and intensity of measured residual nuclei.

Reaction	Residual nucleus	Half life	Energy of γ-ray (MeV)	γ-intensity
$^{27}Al(n,\alpha)$	^{24}Na	15.02h	1.368	99.994%
$^{58}Ni(n,p)$	^{58}Co	70.916d	0.811	99.45%
$^{60}Ni(n,p)$	^{60}Co	5.271y	1.173	99.87%
$^{62}Ni(n,\alpha)$	^{59}Fe	44.496d	1.099	56.5%

Table 2
Results of cross section measurements for $^{58}Ni(n,p)$, $^{60}Ni(n,p)$ and $Ni^{62}(n,\alpha)$.

Cross section (mb) \ Reaction / Energy (MeV)	$^{58}Ni(n,p)$	$^{60}Ni(n,p)$	$^{62}Ni(n,\alpha)$
13.60±0.13	461.0±14.9		
14.09±0.14	366.0±9.2	165.0±4.6	20.4±1.2
14.60±0.22	331.0±13.9		
14.77±0.28	285.0±10.2	142.0±4.5	25.0±1.6
14.81±0.30	290.0±8.2		
15.37±0.44	260.0±9.1		
16.42±0.60	199.0±7.1		
17.77±0.18	166.0±9.4		

reference for neutron fluence rate measurement.

13.6-17.8 MeV neutrons from $T(d,n)^4He$ reaction were used in the measurement, deuteron beam was supplied by cascade accelerator and Van de Graaff accelerator. For the former the mean energy of deuterons was 110 keV, beam current and the diameter of beam spot was about 45 μA and 0.8 cm, respectively, while for the latter the mean energies of deuterons were 1.22 MeV, 1.72 MeV and 1.97 Mev, beam current and the diameter of beam spot was about 15 μA and 0.3 cm, respectively. Target samples were metallic disks with a diameter of about 20 mm that consist of natural isotopic components. Their purity was better than 99.9%. The Al foil thickness was about 10 μm, its mass was about 70 mg. The mass and thickness of Ni foil was usually 500 mg and 0.2 mm, respectively, but a thicker Ni disk with 5400-mg mass and 2-mm thickness was used in the measurement at energy points of 14.09 and 14.77 MeV for obtaining higher γ-counting rate from $^{62}Ni(n,\alpha)$ and $^{60}Ni(n,p)$ reactions. In irradiation, Ni foil was sandwiched between two Al foils, the sample-source distance was 2-4 cm, and the target and beam were at angles of 0°, 20°, 30°, 45° and 120° around the neutron source. In the energy range of 13-15 MeV, neutrons from cascade accelerator were used for irradiation because it gives higher neutron fluence rate, while in the range above 15 MeV those from Van de Graaff were used. The samples were irradiated about 10 hours. Neutron fluence rate was monitored by Au-Si surface-barrier detector on the cascade accelerator and by long counter on the Van de Graaff accelerator.

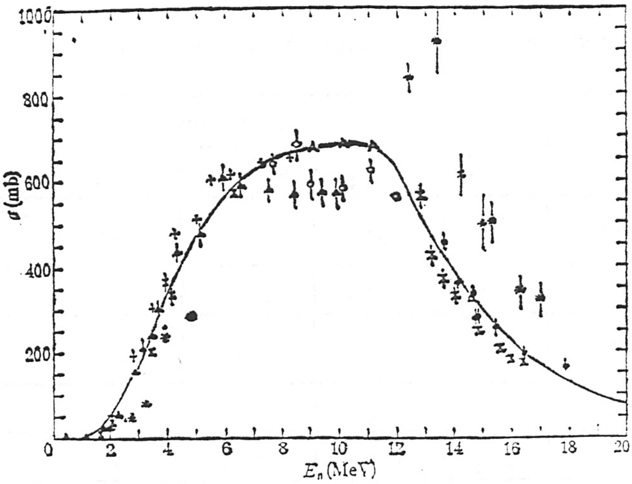

Fig. 1

Cross section of ^{58}Ni(n,p)$^{58m+g}$Co reaction given by different authors. + (62) J. F. Barry [13]; * (68) P. Decowski [14]; ⧖ (71) A. Paulsen [15]; △ (75) D. L. Smith [16]; □ (77) Huang Jianzhou [17]; ◇ (85) Fan Peiguo [18]; ○ (89) H. Vonach [19]; ● (89) this work; ▵ theorical calculation; — evaluation value.

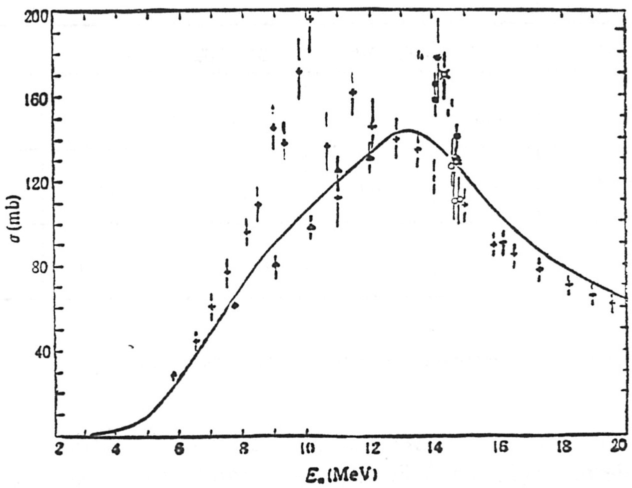

Fig. 2

Cross section of ^{60}Ni(n,p)^{60}Co reaction given by different authors. + (67) A. Paulsen [20]; ✡ (73) J. D. Hemingway [21]; * (74) G. N. Moslov [22]; ⌑ (75) V. Weigel [23]; ⊖ (77) N. I. Molla [24]; ○ (79) E. W. Lees [25]; △ (85) B. M. Bahal [26]; □ (86) N. I. Molla [27]; 1 (89) Wang Yongchang [28]; ⍏ (89) H. Vonach [19]; ● (89) this work; — informal evaluation.

Table 3
Main uncertainty contributions and correlation coefficients in cross section measurement for ^{58}Ni(n,p),^{60}Ni(n,p) and ^{62}Ni(n,α).

Item of contribution	Symbol	Uncertainty (%)			Correlation coefficient of contribution M
		^{58}Ni(n,p)	^{60}Ni(n,p)	^{62}Ni(n,α)	
γ-counting statistics for residual nucleus	ΔN_{γ}	1.1-3.9	1.4-1.8	2.4-3.8	0
γ-counting statistics for ^{24}Na	$\Delta N_{\gamma 0}$	1.0-3.0	1.2-1.7	1.2-1.7	0
γ-detection efficiency for residual nucleus	$\Delta \varepsilon$	1.0	1.0	3.8	1
γ-detection efficiency for ^{24}Na	$\Delta \varepsilon_0$	1.0	1.0	1.0	1
Cross section of ^{27}Al(n,α)	$\Delta \sigma_0$	0.5-2.1	0.5-3.1	0.5-3.1	a
Residual nucleus γ-self-absorption correction in sample	Δfs	0.1	1.0	1.0	1
^{24}Na γ-self-absorption correction in sample	Δfs_0	0.1	0.1	0.1	1
Fluctuation of correction neutron fluence rate for residual nucleus	ΔK	0.07-0.09	0.05-0.09	0.05-0.09	0-1
Fluctuation of correction neutron fluence rate for ^{24}Na	ΔK_0	0.15-0.3	0.1-0.3	0.1-0.3	0-1
Number of target nuclei	ΔM	0.003-0.006	0.003-0.006	0.003-0.006	0.5
Number of target nuclei of Al	ΔM_0	0.017-0.02	0.017-0.02	0.017-0.02	0.5
Scattering correction of neotrons in target head	$\Delta S'$	0.3	0.1	0.2	0-1
Scattering correction of neotrons in sample	ΔS_0	0.1	0.1	0.1	0-1
Error due to neutron energy spread	ΔE	1.7-5.6	0.1-0.7	3.0-8.0	0-1

Note: The correlation coefficient given by S. Tagesen [1].

After irradiation samples were cooled for a period, and then γ-rays from residual nuclei were measured with a Ge(Li) γ-detector (136 cm^3). For a few Al samples γ-rays from residual nuclei were also measured with NaI(Tl) detector with both diameter and height of 80 mm. The detectors were calibrated with reference sources covering the energy range of 0.1-1.5 MeV, and the efficiency curve was calculated with the least square method. For ^{58}Ni(n,p) reaction the cooling time was longer than 5 days and the measurement lasted from 0.5 to 5 hours. For ^{60}Ni(n,p) and ^{62}Ni(n,α) reactions, the cooling time was longer than 6 days and the measurement lasted longer than 9 hours.

The residual nuclei measured in this experiment, their half-life [2], γ-ray intensity and γ-ray energy [3] are listed in Table 1.

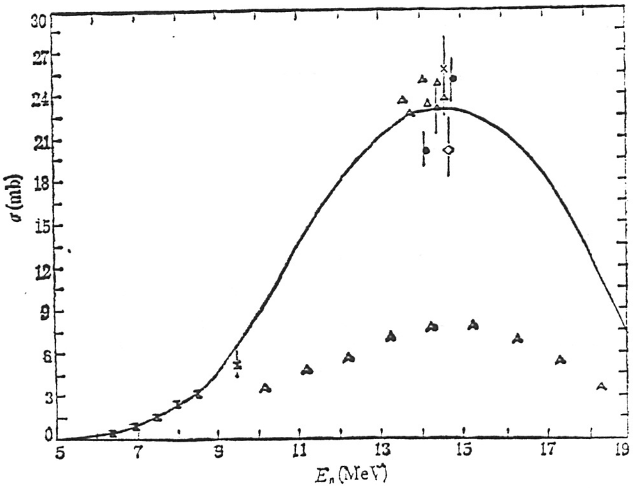

Fig. 3

Cross section of $^{62}Ni(n,\alpha)^{59}Fe$ reaction given by different authors. ✢ (75) V. Weigel [23]; × (78) K. Fukuda [29]; ◇ (79) E. W. Lees [25]; ⧗ (84) S. M. Qaim [30]; △ (89) Wang Yongchang [28]; ● (89) this work; ⩓ theorial calculation; — informal evaluation.

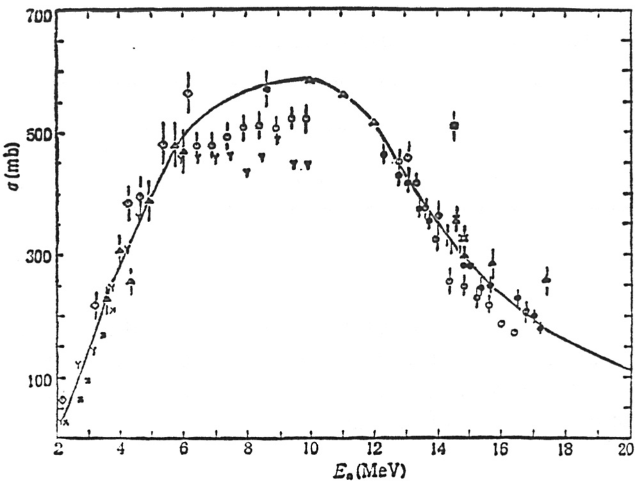

Fig. 4

Cross section of $^{54}Fe(n,p)^{54}Mn$ reaction given by different authors. ✢ (65) E. E. Carroll [31]; × (65) A. Lauber [32]; ◇ (65) S. R. Salisbury [33]; ⊸ (67) P.V. Rao [34]; ▲ (69) R. C. Barrall [35]; ✡ (72) J. J. Singh [36]; Y (75) D. L. Smith [16]; ⧗ (78) K. Fukuda [29]; ⌘ (78) I. Garlea [37]; ⦶ (79) A. Paulsen [20]; ● (82) Lu Hanlin [38]; + (85) B. M. Bahal [26]; ⩓ theorial calculation; — informal evaluation.

Table 4

Relative covariance matrix of cross section error for $^{58}Ni(n,p)$.

	1	2	3	4	5	6	7	8	E_n(MeV)
1	1.00								13.60
2	0.36	1.00							14.09
3	0.23	0.46	1.00						14.60
4	0.27	0.60	0.43	1.00					14.77
5	0.32	0.53	0.40	0.58	1.00				14.81
6	0.21	0.34	0.24	0.33	0.35	1.00			15.37
7	0.20	0.30	0.21	0.29	0.31	0.23	1.00		16.42
8	0.11	0.16	0.12	0.14	0.16	0.13	0.20	1.00	17.77

Table 5

Relative covariance matrix of cross section error for $^{60}Ni(n,p)$.

	1	2	E_n(MeV)
1	1.00		14.09
2	0.36	1.00	14.77

Table 6

Relative covariance matrix of cross section error for $^{62}Ni(n,\alpha)$.

	1	2	E_n(MeV)
1	1.00		14.09
2	0.50	1.00	14.77

From the measured γ-spectrum, counting rates under the concerned total energy peaks were obtained. After corrections in the detector efficiency, cascade effect, self-absorption of γ-ray in sample, γ-intensity, neutron scattering in the sample and target head, and fluctuation of neutron fluence rate, we obtained the cross section σ of the reaction

$$\sigma = \sigma_0 \frac{N_\gamma \cdot e^{\lambda t} \cdot M_0 \cdot \varepsilon_0 \cdot g_0 \cdot I_0 \cdot f_0 \cdot K_0 \cdot S_0 \cdot S_0'(1 - e^{-\lambda_0 T})}{N_{\gamma_0} \cdot e^{\lambda_0 t_0} \cdot M \cdot \varepsilon \cdot g \cdot I \cdot f \cdot K \cdot S \cdot S'(1 - e^{-\lambda T})}, \quad (1)$$

where suffix 0 represents corresponding terms concerning the relative reference Al foil. The meanings of other symbols are: σ — cross section; N_γ — counting rate under total energy peak of the measured characteristic γ-ray; λ — decay constant of the residual nucleus; t — cooling time; M — nucleus number of the sample; ε — detection efficiency for the total energy peak; g — correction for cascade decay; I — γ-ray intensity; f — correction for γ-ray self-absorption in the sample; K — neutron fluence rate fluctuation factor; S — scattering correction of neutrons in the sample; S' — scattering

Table 7
Evaluation of ^{58}Ni(n,p)$^{58m+g}$Co.

Neutron energy (MeV)	Reaction cross section (mb)	Neutron energy (MeV)	Reaction cross section (mb)
0.5	0.02±0.02	10.17	684.0±68.4
1.5	16.4±0.4	11.19	678.0±67.8
2.0	62.7±1.6	12.0	630.6±25.2
3.0	176.0±4.4	13.0	492.4±13.1
3.5	263.8±4.4	14.0	381.4±10.2
4.0	344.6±5.7	14.7	316.8±5.7
4.5	415.9±6.9	15.0	292.8±7.8
5.0	477.0±7.9	16.0	222.6±8.9
5.5	528.1±17.6	17.0	168.1±6.7
6.0	569.8±19.0	18.0	126.6±5.1
7.0	627.9±20.9	19.0	96.3±9.6
8.0	657.4±21.9	20.0	75.6±7.6
9.155	675.0±67.5		

Table 8
Evaluation (informal) of ^{60}Ni(n,p)^{60}Co.

Neutron energy (MeV)	Reaction cross section (mb)	Neutron energy (MeV)	Reaction cross section (mb)
4.0	0.06±0.06	12.5	139.5±13.9
4.5	2.3±1.2	13.0	143.2±10.0
5.0	9.3±0.9	14.0	139.0±4.9
6.0	29.7±2.1	14.7	130.3±3.4
7.0	52.1±3.8	15.0	125.8±4.4
8.0	72.8±5.3	16.0	104.8±7.4
9.0	90.7±6.6	17.0	92.3±6.5
10.0	105.1±10.5	18.0	81.7±5.7
11.0	116.1±11.6	19.0	72.7±5.1
12.0	130.3±13.0	20.0	65.0±4.6

correction of neutrons in the target head; and T — total time of irradiation.

The neutron fluence rate fluctuation factor is

$$K = \left[\sum_{i=1}^{l} \phi_i (1 - e^{-\lambda \Delta t_i}) e^{-\lambda T_i}\right] / \Phi (1 - e^{-\lambda T}).$$

where l is the number of time intervals included in the total time of irradiation; Δt_i is the i-th time interval; T_i is the time from interval Δt_i to the end of the irradiation; ϕ_i is relative neutron fluence rate in interval Δt_i and ϕ is the average relative neutron fluence rate over T.

3. RESULTS OF THE EXPERIMENT

With to Eq. (1) we can obtain cross sections from the measured quantities for ^{58}Ni(n,p),

Table 9
Evaluation (informal) of $^{62}Ni(n,\alpha)^{59}Fe$.

Neutron energy (MeV)	Reaction cross section (mb)	Neutron energy (MeV)	Reaction cross section (mb)
5.0	0	13.5	21.1±4.2
6.5	0.46±0.057	14.0	22.8±1.1
7.0	0.93±0.12	14.5	23.1±1.1
7.5	1.6±2.0	15.0	22.9±1.1
8.0	2.5±0.31	15.5	22.4±4.5
9.0	4.7±0.94	16.0	21.4±4.3
10.0	8.9±1.8	17.0	18.2±3.6
11.0	13.7±2.7	18.0	13.4±2.7
12.0	18.0±3.6	19.0	7.4±1.5
13.0	21.1±4.2		

Table 10
Evaluation of $^{54}Fe(n,p)^{54}Mn$.

Neutron energy (MeV)	Reaction cross section (mb)	Neutron energy (MeV)	Reaction cross section (mb)
2.0	14.4±1.02	12.0	516.7±77.5
2.5	64.8±4.6	12.5	468.7±23.4
3.0	143.4±10.2	13.0	424.5±21.2
3.5	214.7±15.2	13.5	384.5±19.2
4.0	280.0±19.9	14.0	348.3±10.4
4.5	340.5±24.2	14.5	315.5±9.5
5.0	396.7±28.2	15.0	285.8±8.6
5.5	449.3±31.9	15.5	259.0±9.1
6.0	498.8±35.4	16.0	234.8±8.2
6.5	523.8±37.2	16.5	213.0±7.4
7.0	543.1±81.5	17.0	193.5±6.8
8.5	577.1±86.6	17.5	175.9±6.2
9.0	582.5±87.4	18.0	160.3±5.6
9.5	585.7±87.8	18.5	146.4±5.1
10.0	587.0±88.1	19.0	134.1±4.7
10.5	586.7±88.0	19.5	123.3±4.3
11.0	561.4±84.2	20.0	113.9±4.0

$^{60}Ni(n,p)$ and $^{62}Ni(n,\alpha)$ reactions in the energy range of 13.6-17.8 Mev. The results are listed in Table 2.

Contributions of the various uncertainties for each reaction and correlation coefficient of the uncertainty contributions at each energy point are listed in Table 3.

It can be seen from Eq. (1) that reaction cross section σ_i corresponding to incident neutron energy E_i is a function of parameters σ_0, N_γ, M, ε, g, I, f, K, S, S', $N_{\gamma 0}$, M_0, ε_0, g_0, I_0, f_0, K_0, S_0 and S'_0. Suppose that there are L parameters, the error of l-th parameter is e_{il}, for incident neutron energy E_j the error of corresponding l-th parameter is e_{jl} and correlation coefficient is M_{ijl}, covariance matrix of σ_i and σ_j is [4]

$$V_{ij} = \sum_{l=1}^{L} M_{ijl} e_{il} e_{jl} (i,j = 1,\cdots n), \tag{2}$$

where n is the member of energy points at which measurement is made. For an energy point E_i, the error of cross section is

$$\Delta\sigma_i = \sqrt{V_{ii}}, \tag{3}$$

From Eqs.(2) and (3) relative covariance matrix of cross section error can be calculated:

$$C_{ij} = V_{ij} / \Delta\sigma_i \cdot \Delta\sigma_j.$$

Covariance matrices of cross section errors for these three reactions are given in Tables 4, 5 and 6.

4. THEORITICAL CALCULATION

In order to make up for the data that were missing due to a lack of some energy points, we made theoretical calculation for these points with HFTT program [5] in the evaluation. The program was based on a statistical theory which includes emission before equilibrium [6]; the exciton model was used for emission before equilibrium, while the evaporation model was used for emission after equilibrium. The following parameters were used in the calculation: number of initial excitons n_0 = 3(2p,1h); adjustable parameter of exciton transition matrix K = 490 for ^{58}Ni, K = 130 for ^{60}Ni, K = 700 for ^{62}Ni and K = 190 for ^{54}Fe. The density of energy level was take from Gilbet-Cameron formula [7]. The parameter of optical potential was taken from recommended value of Becchetti [8] for n and p, from that of Perey [9] for d, t and ^{3}He, and from that of Mefadden [10] for ^{4}He.

The theoretical value is much lower than the experimental value for the cross section of ^{62}Ni(n,α) reaction. A possible reason for this is that in the theoretical model we used, only the particles above Fermi surface were taken into account for α particle formation factor, but we did not consider that particles on and under Fermi surface could also form α particles [11]. The agreement between theoretical and experimental data will be improved if this factor is taken into account. The problem needs to be solved by further work.

5. DATA EVALUATION

We evaluated experimental data of cross sections which we had collected for ^{58}Ni(n,p), ^{60}Ni(n,p), ^{62}Ni(n,α) and ^{54}Fe(n,p), and made recommendations. Corrections were made on the relative standard cross section, γ-ray intensity and half-life for the collected data. They were normalized at the energy point of 14.7 MeV, and different weight was given to the data of different authors according to their experimental error. Then excitation curve was calculated for the energy range from threshold to 20 MeV.

^{58}Ni(n,p)$^{58m+g}$Co. For this reaction there are more data in the energy range of threshold-6 MeV with good agreement, and the results of our evaluation are consistent with the recommendations in IRDF(1982) [12]. There are fewer data in the range of 6-12 MeV. The only data from Smith [16], Fan [18] and Barry [13] have discrepancy of up to 20%. Therefore, we determined the recommanded value for this range mainly according to the theoretically calculated result. The recommanded value of ours is 20% higher than that of IRDE [12], but it agrees with the data of Fan [18] and Barry [13]. Recently published data show that the results given by Kornilor [39] and Wagner [40] agree with our evaluation, while the data given by Vonach [19] are between those in ENDF/B-V and ours.

In general, the data measured in the past a few years in the range of 12-20 MeV are lower than before, which is also shown by our measurement. Since there are many measured points around 14.7 MeV, we first determined the recommended value at 14.7 MeV carefully in the evaluation, then normalized the data from different authors at this point and gave recommended values in the range of 12-20 MeV by fitting.

^{60}Ni(n,p)^{60}Co. There are few data for this reaction. Most of them are the measured results from Paulsen [20], and other authors' data are mostly around 14 MeV with large discrepancies. The recently published measurement result of Vonach [19] showed a large difference from that of Paulsen [20] in the energy range of 7.7-12 MeV. Therefore, it seems too early for us to give a formal recommended value, and experimental data in more detail are necessary. We gave an informal recommended value with the aid of theoretical calculation and average value in the range of 14.5 MeV.

^{62}Ni(n,α)^{59}Fe. There are very few measured data for this reaction because the low isotopic abundance of ^{62}Ni causes difficulties in measurement. We have collected only 10 datum points from five authors besides two of ours, among which the highest energy point (14.77 MeV) was measured by us. The values given by theoretical calculation for (n,α) reaction, as mentioned above, tend to be low. These facts make evaluation difficult, hence we can only give a rough curve of excitation function. In the evaluation the recommended values in the range of threshold -- 9 MeV are based on the measurement of Qaim [30], while around 14 MeV we determined the recommended value at 14.7 MeV according to the measurement made by other authors and ourselves. The recommended values in other energy ranges were determined by referring to the tendency of theoretical curve and the absolute value at 14.7 MeV. Available experimental data for this reaction are not enough for evaluation at present; the error in evaluation is large and improvement is expected.

^{54}Fe(n,p)^{54}Mn. There are more data for this reaction in the energy range of threshold-6 MeV with good consistency. In this range our evaluation result is basically in accord with those of former authors. Considering that the data of Smith [16] in the energy range of 6-10 MeV are low, we determined the recommended values in this range on the basis of the data given by Fan [18] and referred to the result of theoretical calculation. The values are about 20% higher than the recommended data in the ENDF/B-V and approximate to those in BOSPOR [41]. We collected no datum in the energy range of 10-12 MeV and determined recommended value according to theoretical calculation. In the energy range of 12-20 MeV the main data published in last years are from Paulsen [15] and Lu [38]. They are lower than early data measured by Salisbury [33] and Carroll [31] with recoil proton technique. Consequently, our evaluation results are lower than recommanded values in BOSPOR [41].

Evaluation results for these four reactions are shown in Tables 7-10.

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