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MEASUREMENT OF THERMAL NEUTRON CAPTURE CROSS SECTION AND RESONANCE INTEGRAL FOR THE ^{45}Sc(n,γ)^{46}Sc REACTION WITH PULSED NEUTRONS

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Abstract. *Thermal neutron capture cross section and resonance integral for the $^{45}Sc(n,\gamma)^{46}Sc$ reaction were measured by the activation method using the $^{197}Au(n,\gamma)^{198}Au$ reaction as a single comparator. The high-purity scandium and gold samples with and without Cd cover with the thickness of 0.5 mm were irradiated in a pulsed neutron field of the 100 MeV electron linac of the Pohang Accelerator Laboratory (PAL). The induced activities in the activated foils were measured with a well calibrated HPGe detector. In order to improve the accuracy of the experimental results the effect of the non-ideal epithermal spectrum was taken in to account by determining the neutron spectrum shape factor (α), and the corrections for the thermal (G_{th}) and the resonance (G_{epi}) neutron self-shielding effects, the γ-ray attenuation (F_g) and the γ-ray coincidence summing effect were made. The thermal neutron cross-section for the $^{45}Sc(n,\gamma)^{46}Sc$ reaction has been determined to be σ_o = 27.6 $\pm$ 0.8 barn. By assuming the cadmium cut-off energy of 0.55 eV, the resonance integral for the $^{45}Sc(n,\gamma)^{46}Sc$ reaction has been determined to be $I_o = 12.7 \pm 0.7$ barn. The present results are compared with the reference data and discussed.*

Keywords: thernal neutron cross-section; resonance integral; $^{45}Sc(n,\gamma)^{46}Sc$ reaction; gold monitor; electron linac; Cd-ratio method.

I. INTRODUCTION

The thermal neutron capture cross sections and resonance integrals are of great importance both in fundamental nuclear research and in a various fields of applications [1, 2]. Generally, the neutrons are obtained from the isotopic neutron sources or nuclear reactors with continue neutron spectrum. In this work we used the pulsed neutrons produced from the Pohang neutron facility (PNF) based on the 100 MeV electron linac. The thermal neutron capture cross section and resonance integral for the ^{45}Sc(n,γ)^{46}Sc reaction have been measured by using the ^{197}Au(n,γ)^{198}Au reaction as a single comparator. In recent years similar measurements have been carried out at this neutron facility [3–5]. Scandium is used increasingly in aluminum alloys and the ^{46}Sc radioactive

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isotope is used as a tracing agent in refinery crackers for crude oil [6] as well as in biological and medical studies [7]. Therefore, the accurate knowledge of the ^{45}Sc(n,γ)^{46}Sc reaction cross section is helpful in the production of the medically important radioactive isotope. In literature we have found 12 experimental data [8–19] and 10 evaluation data [20–24] for the thermal neutron capture cross sections as well as 6 experimental data [9,17,19,25,27] and 9 evaluation data [20–22,24,26] for the resonance integral of the ^{45}Sc(n,γ)^{46}Sc reaction. The existing experimental data have been measured before 1985 and the deviations between them are rather large, namely the experimental thermal neutron capture cross sections are varied from 22$\pm$4.4 barn (Seren et al.) to 31.8$\pm$1.6 barn (Harris et al.), with the deviation of 44.54%, and the resonance integrals are varied from 10.7$\pm$0.9 barn (Steinnes) to 14.2 barn (Alian et al.), with the deviation of 32.71%, respectively. In this work, an attempt has been made to provide the new data with the possible highest accuracy. For this aim, the activation measurement was performed by using the pulsed neutrons produced from the electron linac in combination with the Cd-ratio method. In addition, the effects of non-ideal epithermal spectrum, γ-ray attenuation, true coincidence summing of cascading γ-rays, thermal neutron and resonance neutron self-shielding were taken into account during the experimental analysis.

II. EXPERIMENTS

High purity natural samarium metallic foils (99.81%) were used as the activation samples. The Au (99.95%) and In (99.95%) metallic foils were used as the comparator reactions and the neutron flux monitors, respectively. The diameter of all foil samples is 12.7 mm and the thickness is 0.127 mm for Sc, 0.03 mm for Au and 0.05 mm for In, respectively.

The pulsed neutrons were produced by bombarding the accelerated electrons into the tantalum (Ta) target. The Ta target is composed of ten Ta plates with a diameter of 4.9 cm and an effective thickness of 7.4 cm. There was a 0.15 cm water gap between Ta plates in order to cool the target effectively. The housing of the target was made of titanium. The electron beam impinges upon a water-cooled tantalum target and produce bremsstrahlung radiation, which in turn generates so-called photoneutrons via (γ,xn) reactions. The Ta target was set at the center of a cylindrical water moderator made by an aluminum cylinder with a thickness of 0.5 cm, a diameter of 30 cm, and a height of 30 cm. The characteristics of the PNF are described elsewhere [28–32].

The photo-neutrons produced in the giant dipole resonance region consist of a large portion of evaporated neutrons and a small fraction of directly emitted neutrons which dominated at high energies. The neutrons produced in the Ta target without water moderator have a Maxwellian energy distribution with a nuclear temperature of 0.45 MeV. The estimated neutron yield per kW of beam power for electron energies above 50 MeV at the Ta target is about 1.9×10^{12}n/s [33], which is consistent with the calculated value based on Swanson's formula, 1.2×10^{11} $Z^{0.66}$, where Z is the atomic number of the target material [35]. The total neutron yield per kW of beam power was also measured by using the multiple-foil technique and found $(2.30\pm0.28)\times10^{12}$n/s [34]. The neutron energy spectrum with the water moderator is shifted to lower energy region because of the effect of moderation by water. To increase the thermal neutrons in this facility, we have used water to a level of 3-5 cm above the Ta target surface [33]. In this experiment the water level was 5 cm above the target surface. The distributions of neutrons with and without water moderator were described elsewhere [33,34].

In order to measure the thermal neutron cross section and the resonance integral for the ^{45}Sc(n,γ)^{46}Sc reaction by activation method using the ^{197}Au(n,γ)^{198}Au reaction as a single comparator, the natural Sc, Au (with and without a Cd cover of 0.5 mm) and In foils were placed side by side and irradiated simultaneously. The sample holder was placed on the upper surface of the water moderator, 11 cm above the surface of the Ta target, and in parallel with the axis of the Ta target as shown in Fig. 1, where Sc(Cd) and Au(Cd) denote the activation foils covered with a 0.5-mm thick Cd. The irradiation time was 4 hrs. The nuclear reactions of interest are ^{45}Sc(n,γ)^{46}Sc, ^{197}Au(n,γ)^{198}Au, and ^{115}In(n,γ)^{116m}In. Their decay data together with uncertainties (in parenthesis) are given in Table 1 [36]. The neutron fluxes exposed to Sc and Au samples were extrapolated from the measured activities of In monitors.

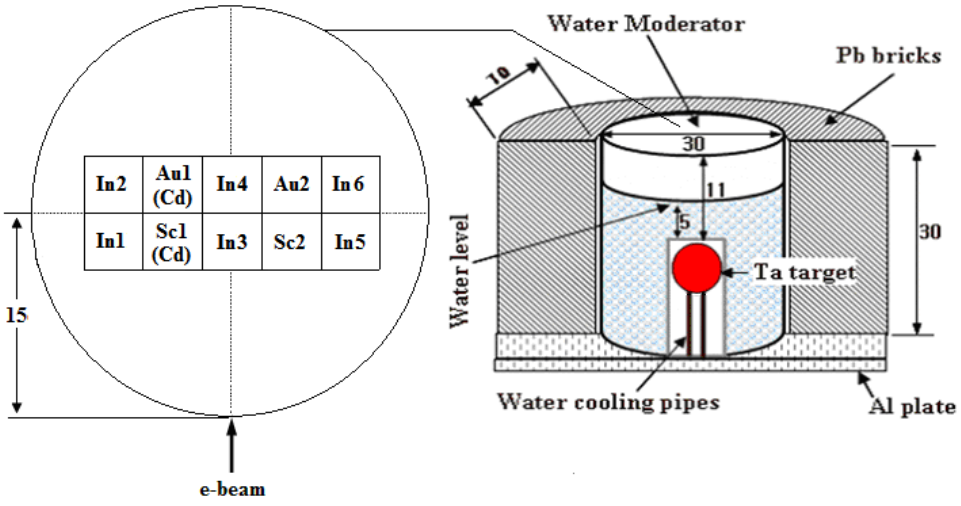

Fig. 1. Configuration of the pulsed neutron source based on the Ta target and water moderator and the arrangement of the activation foils. The numbers in this figure refer to dimension in cm.

Table 1. Nuclear reactions and main decay data [36]

Nuclear reaction	Half-life, $T_{1/2}$	Main gamma - rays		Isotopic abundance (%)
		Energy (keV)	Intensity (%)	
^{45}Sc(n,γ)^{46}Sc	83.79 d (4)	889.277 1120.545	99.984 (10) 99.987(10)	100
^{197}Au(n,γ)^{198}Au	2.69517 d (21)	411.802 675.884	95.58 0.084 (3)	100
^{115}In(n,γ)^{116m}In	54.41 min (17)	416.86 1097.3 1293.545	27.7 (12) 56.2 (11) 84.4 (17)	95.7 (2)

The induced activities of the activated samples were measured by using a well calibrated HPGe detector (Canberra GC2018) with a diameter of 59.2 mm and a thickness of 30 mm coupled to a PC-based multichannel analyzer. The energy resolution of the detector was 1.8 keV full width at half maximum (FWHM) at the 1332.5-keV peak of ^{60}Co. The detection efficiency is 20% at 1332.5 keV relative to a 3" diameter $\times$ 3" length NaI(Tl) detector. The absolute photopeak efficiencies and total efficiencies of the HPGe detector were calibrated with the standard γ-sources. The details of the measurements and calibrations were described in detail elsewhere [37].

The waiting and counting times were decided based on the half-life of the radioactive isotope and statistics of the γ-ray peak of interest. In order to minimize the uncertainties caused by random coincidence and pile-up effects, we have chosen the appropriate distance from the sample to the detector. For all measurements, the dead time was kept below 2%. The activity of the ^{116m}In and ^{198}Au radioactive isotopes were determined based on the 1293.54 keV (84.4%) and the 411.80 keV (95.58%) γ-peaks, respectively. The activity of the ^{46}Sc was determined using two γ-peaks, the 889.277 keV (99.984%) and 1120.54 keV (99.987%) keV. A typical γ-ray spectrum of the activated scandium foil is shown in Fig. 2. The γ-spectra were analyzed using the Canberra software GENIE 2000.

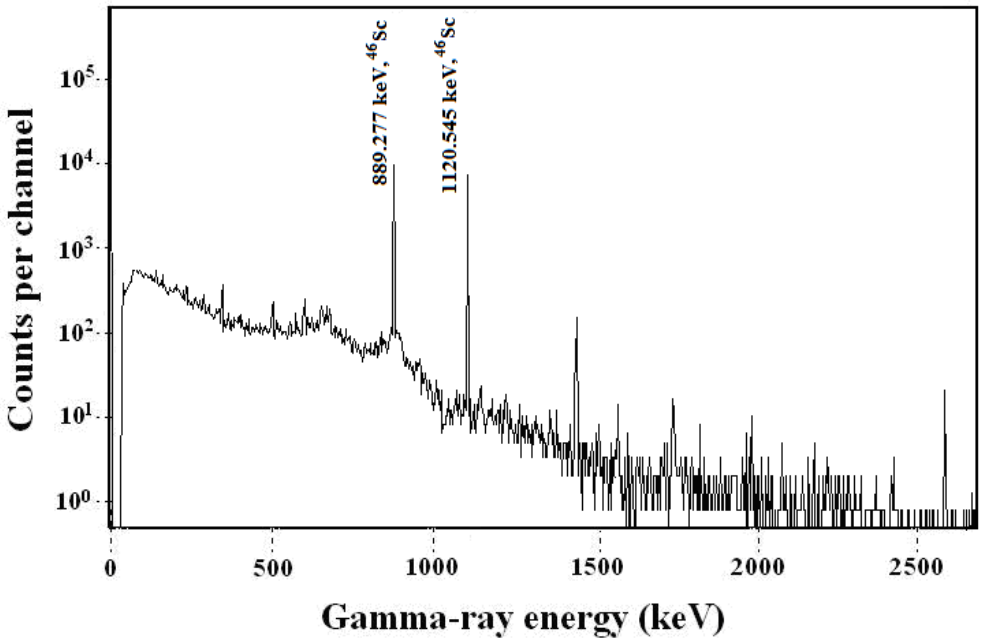

Fig. 2. A typical gamma spectrum of the activated Sc foil with t_i = 4 hr, t_w = 43 hr, and t_m = 1 hr.

III. DATA ANALYSIS

The theory and method for the determination of the thermal neutron capture cross section and resonance integral for the (n,γ) reactions were described in detail elsewhere [3–5], therefore only a brief outline is given here.

III.1. Determination of the reaction rate

The thermal neutron cross section (σ_o) of the ^{45}Sc(n,γ)^{46}Sc reaction was measured using the photo-peak areas of γ-rays at 889.277 keV and 1120.545 keV. The reaction rate (R) of the (n,γ) of the activated foils with *Cd* cover ($R_{x,Cd}$) and without *Cd* cover (R_x) were determined as follows [34]:

$$R_x \quad or \quad R_{x,Cd} \; = \frac{N_{obs}\lambda(1-e^{-\lambda t_{cp}})}{n_o \varepsilon I_\gamma (1-e^{-\lambda\tau})(1-e^{-\lambda t_i})e^{-\lambda t_w}(1-e^{-\lambda t_c})}, \tag{1}$$

where N_{obs} is the net number of counts under the full-energy peak collected during the measuring time t_c, n_o is the number of target nuclei, ε is the detector efficiency, I_γ is the intensity of the γ-ray, λ is the decay constant, t_i is the irradiation time, t_w is the waiting time, τ is the pulse width, and t_{cp} is the cycle period.

III.2. Determination of thermal neutron capture cross section

The thermal neutron cross-section for the ^{45}Sc(n,γ)^{46}Sc reaction, $\sigma_{0,Sc,}$ has been determined relative to that of the ^{197}Au(n,γ)^{198}Au reaction as follows [38]:

$$\sigma_{0,Sc} = \sigma_{0,Au} \times \frac{R_{Sc} - F_{Sc,Cd}R_{Sc,Cd}}{R_{Au} - F_{Au,Cd}R_{Au,Cd}} \times \frac{G_{th,Au}}{G_{th,Sc}} \times \frac{g_{Au}}{g_{Sc}} \tag{2}$$

where $\sigma_{0,Au}$ is the thermal neutron cross-section of the ^{197}Au(n,γ)^{198}Au reaction, R_x and $R_{x,Cd}$ are reaction rates per atom for bare and *Cd*-covered x (*Sc* or *Au*) isotope irradiation, respectively. The cadmium correction factor, $F_{x,cd}$ accounts for the difference in count rate for *Cd* covered and bare samples, and $G_{th,x}$ is the thermal neutron self-shielding factor for x sample. The Westcott factor g_x, correction for departure from $1/v$ cross-section behavior, for the ^{45}Sc(n,γ)^{46}Sc reaction is 1.0002 [39], and that for the ^{197}Au(n,γ)^{198}Au reaction is 1.0054 [39].

III.3. Determination of resonance integral

The resonance integral was measured in non-ideal conditions, where the real neutron spectrum is represented by the semiempirical form 1/E$^{(1+\alpha)}$, where α is an epithermal neutron spectrum shaping factor, which is assumed to be energy independent, the $I_o(\alpha)$ values ought to be used instead of I_o. The conversion of I_o to α-dependent term takes the form [40]:

$$I_0(\alpha) = (1eV)^\alpha \left[\frac{I_0 - 0.429g\sigma_0}{(\bar{E}_r)^\alpha} + \frac{0.429g\sigma_0}{(2\alpha+1)(E_{Cd})^\alpha}\right] \tag{3}$$

where the effective resonance energy, $\bar{E}_r$(eV), as defined by Ryves [41, 42], is required. The term $(I_0 - 0.429g\sigma_0)$ represents the reduced resonance integral, i.e. with the $1/v$ tail subtracted. The literature values of $\bar{E}_r$ are 5.65 eV for ^{197}Au [43] and 5130 eV for ^{45}Sc [43], respectively. The $I_0(\alpha)$ for the ^{45}Sc(n,γ)^{46}Sc reaction was determined relative to that of the ^{197}Au(n,γ)^{198}Au reaction by the following relation [38]:

$$I_{0,Sc}(\alpha) = I_{0,Au}(\alpha) \times \frac{g_{Sc}\sigma_{0,Sc}}{g_{Au}\sigma_{0,Au}} \times \frac{CR_{Au} - F_{Au,Cd}}{CR_{Sc} - F_{Sc,Cd}} \times \frac{G_{epi,Au}}{G_{th,Au}} \times \frac{G_{th,Sc}}{G_{epi,Sc}} \quad , \tag{4}$$

where $CR_x = (R_x/R_{x,Cd})$ are the cadmium ratios, $G_{th,x}$ and $G_{epi,x}$ are the thermal and the epithermal neutron self-shielding factor for x-sample (Sc or *Au*), respectively. The obtained $I_{0,Sc}(\alpha)$ value was converted to $I_{0,Sc}$ by using Eq. (3).

III.4. Correction factors

In the determination of the thermal neutron capture reaction cross section and resonance integral for the ^{45}Sc(n,γ)^{46}Sc reaction, a number of correction factors were made.

The epithermal neutron spectrum shape factor, α at the sample irradiation position was determined based on the dual monitor method using the measured Cd ratios for the ^{197}Au(n,γ)^{198}Au and the ^{186}W(n,γ)^{187}W reactions [4], and it has been found to be 0.067 $\pm$ 0.005. The thermal neutron self-shielding (G_{th}) and epithermal neutron self-shielding (G_{epi}) factors for the gold and scandium samples have been calculated [44,45] as G_{th} = 0.992$\pm$0.005 and G_{epi} = 0.990$\pm$0.007 for Sc, and G_{th} =0.990$\pm$0.007 and G_{epi} = 0.299$\pm$0.005 for Au, respectively. The attenuation factors (F_g) were calculated [4] for each γ-ray of interest and the obtained values are 0.9988 and 0.9990 for the γ-rays of 889.277 keV and 1120.545 keV from ^{46}Sc, as well as 0.994 for the 411.80 keV γ-ray from ^{198}Au, respectively. The measured activity was corrected to zero attenuation by dividing with factor F_g. The 889.277 keV and 1120.545 keV γ-rays emitted from ^{46}Sc are in cascading, therefore the counting loss due to the coincidence summing effect was calculated [46,47]. At a distance of 5 cm between the sample and the HPGe detector the multiplicative factors for the 889.277 keV and 1120.545 keV γ-peaks are 1.022 and 1.025, respectively. The cadmium correction factor for the reactions ^{45}Sc(n,γ)^{46}Sc and ^{197}Au(n,γ)^{198}Au are 1.000 and 1.009 [48], respectively. The main correction factors used for the determination of thermal neutron capture cross-sections and resonance integrals of the investigated nuclear reactions ^{45}Sc(n,γ)^{46}Sc and ^{197}Au(n,γ)^{198}Au are listed in Table 2 [39, 43, 48].

Table 2. Main correction factors for the calculations of thermal neutron capture cross-section and resonance integral.

Nuclear reaction	$\overline{E}_r$,eV [43]	Q_0 [43]	G_{th}	G_{epi}	F_{Cd} [48]	g [39]
^{45}Sc(n,γ)^{46}Sc	5130	0.43	0.992$\pm$0.005	0.990$\pm$0.007	1.000	1.0002
^{197}Au(n,γ)^{198}Au	5.65	15.7	0.990$\pm$0.007	0.299$\pm$0.005	1.009	1.0054

IV. RESULTS AND DISCUSSIONS

The present thermal neutron capture cross-section and the resonance integral for the ^{45}Sc(n,γ)^{46}Sc reaction were found to be $\sigma_{0=}$ 27.6$\pm$0.8 barn and I_0= 12.7$\pm$0.7 barn, respectively. The present results together with reference data are given in Table 3. The main sources of uncertainties for the thermal neutron capture cross-section measurement are due to the detection efficiency (2.2%), statistical error (0.5%) and thermal neutron shelf-shielding factor (0.5%). The main sources of uncertainties for the resonance integral measurement are due to α-shape factor (3.3%), epithermal neutron self-shielding factor (1.6%), reference resonance integral cross section of ^{197}Au (1.81%), and cadmium ratio (1.45%). The total uncertainties for the thermal neutron cross-section and the resonance integral for the ^{45}Sc(n,γ)^{46}Sc reaction have been estimated to be 3.1% and 5.6%, respectively.

The present thermal neutron capture cross section for the ^{45}Sc(n,γ)^{46}Sc reaction $\sigma_0 = 27.6 \pm 0.8$ barn is in good agreement within 1σ level with the reference data measured by Mughabghab (1984), Mannhart (1975), Dilg (1974), and Wolf (1960); within 2σ level with

the data measured by Wilson (1967), and Gleason (1975); and within 3σ level with the data measured by Pattenden (1955). The present result disagrees with old data measured by Seren (1947), Harris (1950), Pomerance (1951), Lyon (1953) by 15.2-20.2%, respectively. The present result is in agreement with almost evaluated values within 1σ level, and within 2σ level with the values evaluated by De Corte (2003) and by Kafala (1997), respectively.

The present resonance integral value I_o = 12.7±0.7 barn at the cadmium cut-off energy of 0.55 eV for the ^{45}Sc(n,γ)^{46}Sc reaction is in agreement within 1σ level with the data measured by Harris (1950) and Van der Linden (1972), Gleason (1975), Mughabghab (1984); and within 3σ level with the data measured by Alian (1973) and by Stainnes (1972), respectively. The present resonance integral result is in agreement within 1σ level with the data evaluated by ENDF/B-VII.1 (2011) and by Atlas (2006); and within 2σ level with all other evaluated values.

Table 3. Thermal neutron capture cross sections and resonance integrals for the ^{45}Sc(n,γ)^{46}Sc reaction

Year	**Authors**	**σ_0(barn)**	**I (barn)**	**Monitor**
2013	This work	27.6 ± 0.8	12.7 ± 0.7	Au
2011	ENDF/B-VII.1 [24]	27.16	12.06	Evaluation
2011	JEFF3.1.2 [24]	27.15	11.83	Evaluation
2011	JENDL4.0 [24]	27.14	11.84	Evaluation
2010	EAF2010 [24]	27.16	11.84	Evaluation
2010	ROSFOND2010 [24]	27.15	11.84	Evaluation
2006	Atlas [24]	27.2±0.2	12.1 ± 0.5	Combination
2003	F. De Corte [23]	26.3 ± 0.16	-	Combination
1997	S.I. Kafala *et al.* [22]	26.4 ± 0.2	11.8 ± 0.4	Combination
1989	F. De Corte, A. Simonits [21]	27.2 ± 0.7	12 ± 4.2	Combination
1987	E. Gryntakis *et al.* [20]	27.2 ± 0.2	11.5 ± 0.5	Combination
1984	S.F. Mughabghab [19]	27.2 ± 0.2	12 ± 0.5	Au
1978	M.Takiue, H.Ighikawwa [18]	23.4 ± 0.4	-	Co
1975	G.Gleason [17]	26 ± 5	12 ± 1.0	Au, Mn
1975	W.Mannhart [16]	27.07 ± 0.17	-	Au
1974	W.Dilg, W.Mannhart [15]	27.54 ± 0.2	-	TOF
1973	A. Alian *et al*. [27]	-	14.2	Co
1972	R. Van der Linden *et al*. [26]	-	13 ± 1	Combination
1972	E. Steinnes [25]	-	10.7± 0.9	Au
1967	W.L.Winlson [14]	26.6 ± 0.5	-	Co
1960	G.Wolf [13]	28.3 ± 0.7	-	Au
1955	N.J.Pattenden [12]	25.5 ± 1	-	TOF
1953	W.S.Lyon [11]	23.3 ± 2.33	-	Mn
1951	H.Pomerance [10]	23 ± 1.15	-	Au
1950	S.P.Harris *et al.* [9]	31.8 ± 1.6	12.6	Au
1947	L.Seren *et al.* [8]	22 ± 4.4	-	Absolute

V. CONCLUSION

The thermal neutron cross section and the resonance integral for the ^{45}Sc(n,γ)^{46}Sc reaction have been measured by the activation method using the ^{197}Au(n,γ)^{198}Au reaction as the single comparator. The activation was performed with the pulsed neutrons produced from the 100 MeV electron linac. In order to improve the accuracy of the experimental results the necessary corrections were made. The obtained results for the thermal neutron cross section and the resonance integral of the ^{45}Sc(n,γ)^{46}Sc reaction are σ_o = 27.6 $\pm$ 0.8 barn and $I_o = 12.7 \pm 0.7$ barn, respectively. The present results are closed to the mean values of the existing reference data. The difference between the present thermal neutron capture cross section and the average value of the existing reference data is 3.9%, and in case of the resonance integral the difference between the two values is 4.5%, respectively.

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