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NEUTRON ACTIVATION CROSS SECTION FOR NI ISOTOPES AT 14.8 MEV

I. Ribansky, J. Kristiak, L. Stoeva and C. Panteleev

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Neutron activation cross section for Ni isotopes at 14.8 MeV+

Ribanský I., Krištiak J.

Institute of Physics EPRC, Slovak Academy of Sciences, CS-84228 Bratislava, Czechoslovakia

and

Stoeva L., Panteleev C.

Institute of Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences, Sofia 1181, Bulgaria

Neutron activation cross sections for Ni isotopes were measured using Ge(Li) γ-ray spectroscopy of the reaction products. The linear least-squares method was used to resolve the interfering reactions. The results obtained are compared with recent experimental and evaluated data, statistical model and semiempirical calculations.

1. INTRODUCTION

Though very many measurements of neutron activation cross sections at ~14 MeV were reported in the literature up to now the new data are still required as documented e.g. by the recent issue of WRENDA 83/84++. There are many

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++World Request List for Nuclear Data, published by IAEA, Vienna 1983.

reasons behind those new requirements. The most important one, we believe, is the fact that the reported cross sections often exhibit large mutual disagreement greatly exceeding quoted uncertainties (see e.g. Csikai [1], Bychkov et al. [2]). For the evaluators the renormalization of different results is very difficult and the reconsideration of quoted uncertainties almost impossible. This makes the significance of such data rather questionable both from the point of view of their practical applications (e.g. fusion reactor design) and theoretical interpretation (e.g. test of models describing the reaction mechanism). It appears that the only way to improve the existing situation is the careful remeasurements of the important neutron activation data using modern experimental techniques, data processing procedures and error estimation methods.

In this paper we present our measurements of the neutron activation cross sections for Ni isotopes with the emphasize to (n,p) and (n,np)+ processes. Such data are needed because the nickel is one of the potential component of the fusion reactor construction materials. Also the comparison of our data with the recent integral and differential measurements can contribute to the identification of the "best" values and the comparison with the theoretical models can shed light on the reaction mechanism or their competence to describe it.

+ The (n,np) symbol stands for the sum of (n,n'p), (n,pn) and (n,d) contributions.

2. EXPERIMENTAL PROCEDURE

The details of the experimental technique and data aquisition system were described in our previous papers [3, 4]. Here we report only the details pertinent to the present experiment.

The samples used were prepared by pressing NiO or pure metal powder into plexiglass containers (ø 16 mm). The enriched samples were supplied by TECHSNADEXPORT, Moscow. All samples were of spectral purity. Their isotopic abundances are listed in table 1. The sample thickness varied from 250 to 600 mg/cm^2.

The required neutrons were produced via $T(d,n)^4He$ reaction and irradiations were performed at zero degrees with respect to deutron beam. The time variation of the neutron yield was monitored by two neutron detectors and was taken into account in the cross section calculations. the present experiment. The covariance matrix method as proposed by Mannhart [10] and Smith [11] was employed for the estimation of total uncertainties. The sources of uncertainties which were taken into account in this work are listed in table 2. All uncertainties quoted in our paper represent one standard deviation.

3. RESULTS AND DISCUSSION

The final results of our measurements are presented in table 3. Those data which are correlated with each other are grouped and the corresponding correlation matrix is given in the last column. The number of independent irra-

Table 1: Isotopic abundances (%) of Ni samples

Sample	Isotope 58	60	61	62	64
Natural Ni	68.27	26.10	1.13	3.59	0.91
^{58}Ni	99.8	0.2	0.05	0.04	0.19
^{60}Ni	0.08	99.9	0.01	0.01	0.01
^{61}Ni	9.2	20.7	66.6	2.9	0.6
^{64}Ni	3.64	2.06	0.25	0.95	93.1

Table 2: The principal sources of uncertainties

Source	Resulting uncertainty (%)
Couting statistics	0.5 - 6
Sample mass	0.1
Isotopic abundances	0.2
γ-ray intensities	0.3 - 2.2
Detector efficiency (FEP)	1.5
Reaction product half-life	0.3 - 3.0
Irradiation counting geometry (position)	0.2
Coincidence summing corrections	0.7
γ-ray selfabsorbtion	0.5
Monitor calibration (including ^{56}Fe(n,p) reference reaction)	2.8

diations is given in the 4th column. The decay data of the reaction products are presented in table 4. Here the last column contains the values of the coincidence summing corrections for the γ-transition used. The numbers in brackets (column 2 and 4) represent the uncertainties in the same format as used by Lederer and Shirley [12].

Table 3: Neutron activation cross sections for Ni isotopes at 14.8 MeV

Reaction	Measured cross section (mb)	Error (%)	No. of measurements	Correlation matrix (%)	
$^{58}Ni(n,2n)^{57}Ni \rightarrow {}^{57}Co$	32.6	8.6	6	100	
$^{58}Ni(n,np)^{57}Co$	467	5.1	4	-28	100
$^{58}Ni(n,p)^{58m}Co$	134.2 a)	4.6	3	100	
$^{58}Ni(n,p)^{58g}Co$	120.4 a)	5.7		-29	100
$^{58}Ni(n,p)^{58m+g}Co$	254.6	2.8			
$^{60}Ni(n,p)^{60m}Co$	50.1	8.4	14	100	
$^{61}Ni(n,np)^{60m}Co$	25.8	3.6		-49	100
$^{61}Ni(n,p)^{61}Co$	66.6	4.7	17		
$^{62}Ni(n,p)^{62m}Co$	11.7 b)	6.8	4	100	
$^{62}Ni(n,p)^{62g}Co$	17.3 b)	6.9		-66	100
$^{62}Ni(n,p)^{62m+g}Co$	29.0	3.1			
$^{64}Ni(n,np)^{63}Co$	0.86	4.7	6		
$^{64}Ni(n,\alpha)^{61}Fe$	4.28	3.7	6		

a) Analysis of the 810.8 keV γ-line decay curve

b) Analysis of the 1129.1 keV, 1163.5 kev and 1173.0 keV γ-line decay curves

3.1 Ni(n,p) reactions

$^{58}Ni(n,p)^{58}Co$ reaction: Both the isomeric (σ_m) and the ground (σ_g) state cross sections were determined in this work by analysing the decay curve of the 811 keV γ-transition.

Table 4: Decay data of reaction products

Reaction product	Half-life	E (keV)	I (%)	Ref.	Coincidence summing correction
^{57}Ni	36.08(9) h	127.2	12.9(9)	a)	1.20
		1377.6	77.9(23)	a)	
^{57}Co	271.73(14)d	122.1	85.68(13)	b)	1.00
		136.5	10.67(13)	b)	1.00
^{58m}Co	9.15(10)h	24.9	100.00(IT)	c)	1.00
^{58g}Co	70.80(8) d	810.8	99.4(3)	a)	1.03
^{60m}Co	10.47(2) m	58.6	2.07(13)(IT)	a)	1.00
^{61}Co	1.650(5)h	67.4	86(3)	c)	1.01
^{62m}Co	13.91(5) m	1163.5	62(2)	c)	1.12
		1173.0	97.9(2)	c)	1.12
^{62g}Co	1.50(4) m	1129.1	13.1(9)	c)	1.11
		1173.0	82.6(6)	c)	1.02
^{63}Co	27.5(3) s	87.1	49.3(15)	c)	1.00
^{61}Fe	5.98(6) m	1027.4	42.7(26)	c)	1.07
		1205.1	43.6(14)	c)	1.01

IT = isomeric transition

a) Ref. [34]

b) Ref. [35]

c) Ref. [12]

Table 5: Comparison between experimental and calculated (n,p) activation cross sections

Reaction	Experimental cross section (mb) a	Calculated cross section (mb) b	c	F_{PEQ}
$^{58}Ni(n,p)$	255 ± 7	290	344	0.16
$^{60}Ni(n,p)$	-	127	121	0.19
$^{61}Ni(n,p)$	67 ± 3	65	74	0.56
$^{62}Ni(n,p)$	29 ± 1	27	46	0.44
$^{64}Ni(n,p)$	-	6	18	0.67

[a] This work

[b] Proequilibrium model

[c] Levkovskii's formula

This reaction was frequently investigated in the past. The measured excitation functions exhibit a large gradient ($\sim$200 mb/MeV) around 14 MeV. We shall therefore compare our results with only those values measured recently at $\sim$14.8 MeV.

Viennot et al. [13] measured the excitation function around 14 MeV. From their results we deduced $\sigma_{tot} = \sigma_m + \sigma_g =$ 269±9 mb which is in agreement with our result. Similarly, from the excitations functions measured by Raics et al. [14] we found σ_{tot}=268±10 mb and the isomeric ratio $\sigma(5^+)/\sigma(2^+) = \sigma_m/\sigma_g = 1.2 \pm 0.1$. Both values agree with our results within the quoted uncertainties. On the other hand the excitation functions measured by Hudson et al. [15] imply σ_{tot}=331±26 mb and σ_m=166±13 at 14.8 MeV. Their σ_{tot} deviates significantly from our value. The same is true for σ_{tot}=338±22 mb measured by Fukuda et al. [16] and σ_{tot}=375±22 mb reported by Qaim and Stöcklin [17] . Similar disagreement is observed for evaluated values 347±5 mb [7] and 410±30 mb [2].

Our σ_{tot} can also be compared with Grimes et al. [18] spectral data. To do so one usually assumes that the emission of the second particle starts immediately at the second particle threshold S_2. The quantity to be compared with the activation cross section is therefore the integral from S_2 to ε_p^{max} (maximum energy of the emitted proton). Considering only nucleon emission S_2=E−min(B_{pn},B_{2p}), where B's are the binding energies of nucleon pairs in ^{59}Ni and E is its excitation energy. In our case B_{2p}=B_{pn}−1.6 MeV [19] and for the corresponding integral we obtained 130±22 mb a value which is too small compared to measured activation cross section. However due to essentially zero reaction cross section for protons with $\varepsilon_p \in (0, 1.6)$ MeV − σ_R (ε_p=1.6 MeV)≈1 mb, Mani et al. [20]−almost no secondary protons are emitted up to the S_{pn} threshold. Starting the integration from S_{pn} we obtained ~250±40 mb which seems to support our result. Should there be, however, any reason why the emission of the second neutron from ^{58}Co to low lying levels of ^{57}Co be hindered the integration should start below S_{pn} and the integral could reach value well above 300 mb as $d\sigma/d\varepsilon \approx 100$ mb/MeV near S_{pn}. Considering the spin distribution of the compound nucleus ^{59}Ni (see discussion bellow) and the level scheme of ^{57}Co [12] we feel it will not happen.

$^{60}Ni(n,p)^{60}Co$ reaction: Due to the long half-life of the ^{60g}Co nucleus we were able to measure only σ_m cross section. Our value is in strikking disagreement with the two recent measurements: 26±3 mb (Viennot et al. [13]) and 95±10 mb (Molla and Qaim [21]) though the same decay data and experimental technique were used. We would like to argue that our value should be closer to the true one.

The recent measurements of $\sigma_{tot} = \sigma_m + \sigma_g$ are concentrated around ~125 mb : 112±12 mb Molla and Qaim [21], 134±11 mb Fukuda et al. [16] and 128±13 mb Lass et al. [22]. These results are also in good agreement with the integral (from S_{pn}) of the experimental $^{60}Ni(n,p)$ proton spectrum (136±20 mb) measured by Grimes et al. [18]. One can therefore expect $\sigma_g \equiv \sigma(5^+) \approx 30$ mb if $\sigma_m \equiv \sigma(2^+) \approx 95$ mb or $\sigma_g \approx 95$ mb if $\sigma_m \approx 30$ mb. This implies $\sigma(5^+)/\sigma(2^+) \approx 0.3$ or 3 accordingly. Both ratios appear to be improbable. Using the transmission coefficients of Mani et al. [20] we have calculated the spin distribution of the compound nucleus. It is centered at ~3.8 $\hbar$ which is just between the spins of the ground state (5^+) and the isomeric state (2^+) of the ^{60}Co nucleus. Because this spin distribution is not appreciably changed after the proton emission we expect the ratio $\sigma(5^+)/\sigma(2^+)$ to be close to 1 as was the case for $^{58}Ni(n,p)$ reaction. Our σ_m does satisfy this expectation.

$^{61}Ni(n,p)^{61}Co$ reaction: The cross section measured in this work disagrees with the recent measurements of Molla and Qaim [21] 95±10 mb and Viennot et al. [13] 93±15 mb. The same holds for the evaluated value 95±10 mb Bychkov et al. [2]. We did not find an indication which value should be more appropriate as in both cases the same experimental technique and decay data were used. We note that the theoretical calculations (see sect. 3.4) seems to support our result.

$^{62}Ni(n,p)^{62}Co$ reaction: Both σ_m and σ_g cross sections were determined in this work. Our σ_m is ~2 times smaller than σ_m=21±2.5 mb measured by Molla and Qaim [21]. Viennot et al. [13] also measured both cross sections and found σ_m=19±2 mb which differ substantially from our value and σ_g=22±2 mb which is close to our result. We have no reasonable explanation for the observed differences.

3.2 Ni(n,np) reactions

$^{58}Ni(n,np)^{57}Co$ reaction: The cross section determined in this work is corrected for the contribution of $^{58}Ni(n,2n)^{57}Ni$ reaction which leads to the production of ^{57}Co nucleus via β^+ decay of ^{57}Ni. Our value is in good agreement with 520±50 mb reported by Qaim [23]. Other value reported by Fukuda et al. [16] (373±29 mb) and Raics et al. [14] (630±27) mb differ substantially from our result.

$^{61}Ni(n,np)^{60}Co$ reaction: Only σ_m was determined in this work due to long half-life of the ^{60g}Co nucleus. Our value turned out to be ~2 times higher than the only recent result reported by Qaim [23] : 13.0±3.0 mb. Surprising fact is that our σ_m for $^{60}Ni(n,p)$ reaction leading to the same ^{60m}Co nucleus was ~2 times smaller than the Jüllich value. This implies that the ratio of the (n,p) and (n,np) isomeric cross sections - as measured at Jüllich - is ~ 4 times higher than our ratio while the sum of those cross sections differs only by 30 %, our being smaller. If we are in error the main source of the observed discrepancy has to be hidden in the linear least-square proceoure which in fact evaluates how much of the observed activity of ^{60m}Co should be attributed to different - in our case two - sources. We found that such an error is statistically highly improbable ($\chi^2_N \approx 1$). We believe that the simultaneous measurements of the (n,p) and (n,np) cross sections is more reliable than separate measurements corrected for the contributions from other interfering reactions. Our belief is based on the fact that σ_m for ^{60}Ni(n,p)^{60m}Co reaction obtained from the irradiations of the samples higly enriched in ^{60}Ni (99,9 %) remained the same when the data from other isotopic mixtures were used in the linear least-square procedure.

^{64}Ni(n,np)^{63}Co reaction: This reaction was also investigated recently at Jülich. Their renormalized activation cross section 3.0±0.4 mb (Qaim [23]) is again in striking disagreement with our result. Though the same decay data were used in both cases the results differ by a factor of 3. We have no explanation for this enormous difference.

3.3. Other n+Ni reactions

^{58}Ni(n,2n)^{57}Ni reaction: This reaction was measured primarily because it interferes with ^{58}Ni(n,np) reaction. Our result is in excelent agreement with the majority of other data measured recently: 35±3 mb Qaim and Molla [24], 25.6±3 mb Hudson et al. [15], 27.5±3 mb Adamski et al. [25], 30±6 mb Molla et al. [26], 35.3±1.4 mb Raics et al.

[14], 34.8±2.1 mb Csikai [27], 40.3±1.5 mb Lu et al. [28] and 39.63±0.52 mb Winkler et al. [29]. The same holds for the evaluated values 35.1 mb [7] and 30 mb Bychkov et al. [2].

^{64}Ni(n,α)^{61}Fe reaction: This reaction was rearly investigated in the past. As far as we know there exists only one activation cross section measurements at 14.8 MeV due to Levkovskii [30]: 5.2±1.2 mb. This value is in agreement with the present result within the quoted uncertainties.

3.4 Comparison with calculated (n,p) cross sections

Now we compare our (n,p) activation cross sections with the preequilibrium statistical model calculations and the predictions based on the semiempirical formula derived by Levkovskii [31].

The preequilibrium model employed in this work is described in detail by Ribanský [32]. It differs from that used in our previous paper (Ribanský and Gmuca [4]) in the following. The model assumes that only those nucleons lying on the last occupied subshells of the target nucleus are involved in the preequilibrium stage of a reaction. The number of excited protons (neutrons) are calculated using the free NN cross section. As a result the hole degrees of freedom are essentially eliminated and the spectra of emitted particles are harder. The only free parameter is the normalization constant for the intranuclear transition rate. In our calculation it was determined from the simultaneous fit to the measured neutron (Hermsdorf et al. [33]) and proton (Grimes et al. [18]) spectra at ~14 MeV for n+Ni reactions.

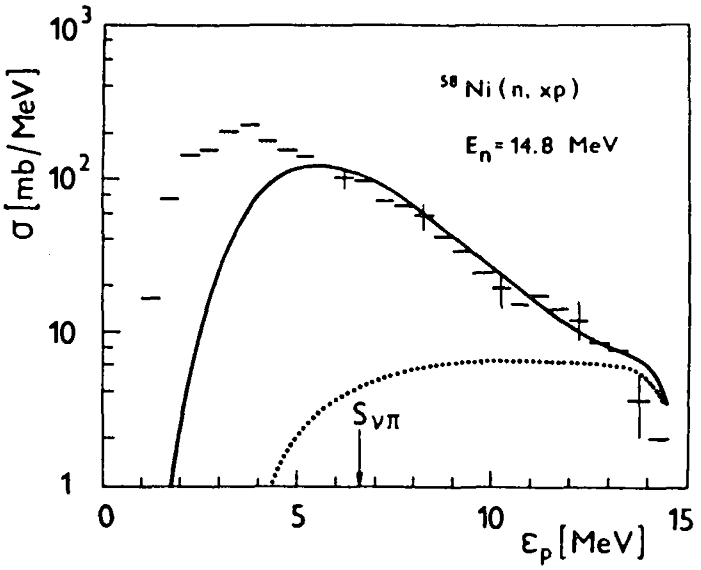

Figure 1. The comparison between the experimental data (histogram) of Grimes et al. (1979) and the calculated proton spectrum (full curve) for ^{58}Ni(n,xp) reaction. The dotted curve represent the preequilibrium part of the proton emission. The arrow indicates the threshold for the second particle emission.

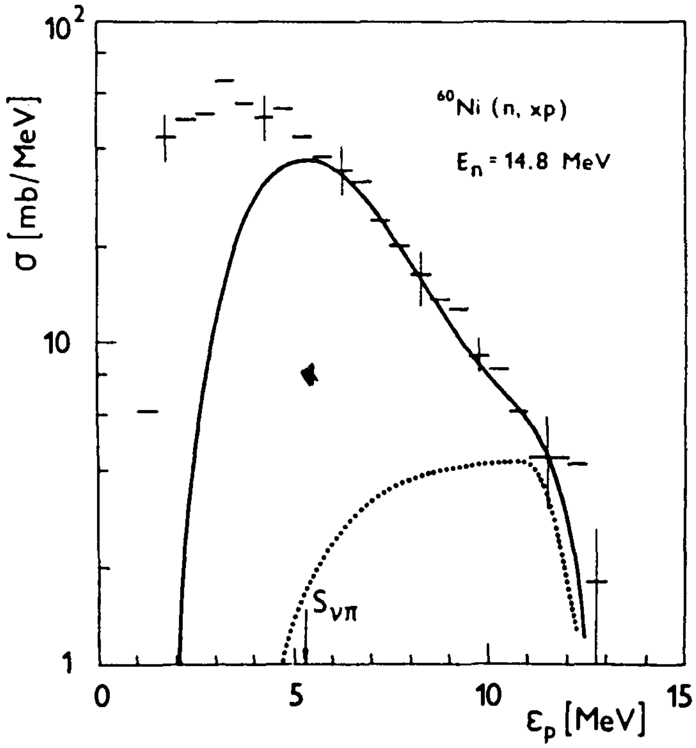

Figure 2. As in figure 1 for ^{60}Ni(n,xp) reaction.

The calculated first emitted proton spectra are compared with the experiment in figs. 1 and 2. The calculations reproduce the experiment very well both in magnitude and shape. Obviously this is essential for the comparison of the calculated activation cross sections and those deduce from the spectral data. In table 5 the comparison between the experimental and calculated (integrals from S_{pn} to ε_p^{max}) (n,p) cross sections are presented (column 2 and 3). The calculated values agree quite well with our measured data giving some confidence to their reliability. The semiempirical predictions (column 4) are in quite good agreement with our theoretical values for the first three reactions. For the last two reactions this difference is larger reaching a factor of 3 for ^{64}Ni(n,p) reaction. This is not surprizing because the Levkovskii's formula is based on the evaporation mechanism and should fail to describe situation where the preequilibrium emission is substantial. The calculated fraction of preequilibrium emission F_{PEQ} (last column) indicates that this is really the case.

It would be very intersting to measure the cross section for ^{64}Ni(n,p) reaction in order to test our prediction. Extrapolation of our experimental data assuming σ_{np}= (-1+exp(0.859 x)) mb, where x= $\varepsilon_p^{max}-S_{pn}$ indicates that $\sigma_{np}(^{64}Ni)$ should indeed be around 6 mb. The recommended value of Bychkov et al. [2] 5±1 mb supports our prediction.

4. CONCLUSION

The neutron activation cross sections for Ni isotopes at 14.6 MeV were measured using Ge(Li) γ-spectroscopy of the reaction products. The linear least-square method was used to resolve (n,p) and (n,np) reactions on the neighbouring isotopes leading to the same reaction product. This method which is based on the use of targets with several different isotopic compositions seems to be more reliable than the separate measurements corrected for the contribution from interfering reactions. An effort was made to treat correctly all errors - we are aware of - associated with the present experiment.

Our measured cross sections were compared with the recent experimental data. A large deviations were observed for (n,p) and (n,np) cross sections. In several cases we found arguments favouring our results. These were however not sufficiently conclusive and, clearly, still new measurements are needed to improve the situation.

The (n,p) cross sections were compared with the calculations based on the statistical preequilibrium model. These calculations support our (n,p) data. The semiempirical predictions were found to overestimate progressively the (n,p) cross sections for the neutron rich Ni isotopes. The large ($\sim$50 %) contribution of the preequilibrium emission seems to be responsible for this trend.

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