Neutron-induced activation cross-sections for fusion technology applications

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Abstract. Activation method was employed to measure the cross section for the $^{58}$Ni(n,p$\alpha$)$^{54}$Mn and $^{63}$Cu(n,p$\alpha$)$^{59}$Fe reactions in the energy range from the threshold up to 21 MeV. The quasi-monoenergetic neutrons in the energy range from 14 to 21 MeV were produced via the $^3$H(d,n)$^4$He reaction at 1, 2, 3 and 4 MeV incident deuteron energies. High purity Ni and Cu samples with natural isotopic composition and dimensions of ø12x5 mm and ø20x5 mm respectively were utilized. The gamma-ray spectrometry using HPGe detector was utilized to measure the activity induced in the sample. The cross sections were determined relative to the standard cross section the $^{27}$Al(n,$\alpha$)$^{24}$Na reaction. The results for the $^{63}$Cu(n,p$\alpha$)$^{59}$Fe reaction cross section were obtained for the first time.

1. Introduction
Neutron-induced reaction cross sections are important for many fields of science and technologies. Activation and transmutation analyses for the high intensity neutron sources require a full set of cross section data comprising all target nuclides that may be present in the materials to be irradiated. Radiation damage is largely governed by the helium and hydrogen production. The radiation hazards and decay heat are associated with the decay of the activated products. Nuclear data are needed for the calculation of the medical radioisotope production as well [1]. Accurate experimental data for neutron-induced activation cross sections are important for the validation and improvement of the nuclear models.

Nuclear fusion is recognised as a long-term energy source. The most important initiative on fusion research is currently the ITER project. Safety and environmental issues are of great importance in the continuing development of the nuclear power plants. In this respect accurate, complete and reliable neutron-induced activation cross-section database is needed.

Experimental cross section data for the multiple charged particle emission reactions are scarce. There are very few datasets for the studies (n,p+alpha) reactions available in Experimental Nuclear Reaction Data EXFOR database [2].

In this work, we report the finalized results for the (n,p$\alpha$) reaction cross sections on $^{58}$Ni and $^{63}$Cu isotopes in the energy range from 14 to 21 MeV. The experimental procedure [3] is presented in section 2. The section 3 describes the data and uncertainty analysis. The results from this work are presented and compared with the measurement by other authors, evaluated data [4] and TENDL library [5] in section 4.
2. Experimental method

Disc-shaped high purity natural nickel (ø12 x 5 mm) and copper (ø20 x 5 mm) samples supplied by Goodfellow were employed in the measurements. The mass of the nickel and copper samples were about 5 g and 14 g, respectively. Due to the low cross sections in the studied energy range the interference from the reactions on the impurities, present in the samples, needed to be taken into account. The sample impurities (ppm) provided by the supplier for the nickel material are: Ag 8, Al 15, Au 15, B 20, Ca 20, Co 5000, Cr 30, Cu 80, Fe 40, Mg 8, Mn 100, Pb 15, Si 80, Zn 10 and for the copper are: Ag 5, Al 5, B 8, Bi 10, Ca 25, Cd 2, Cr 2, Fe 2, K 100, Mg 1, Mn 1, Na 2, Pb 100, Si 4. Aluminium and niobium monitor foils of the same diameter and 0.1 mm thickness were attached at the both sides of the Ni and Cu samples.

2.1. Irradiations

The irradiations were carried out at the 7 MV Van de Graaff accelerator. Quasi mono-energetic neutrons were produced via the \( ^3\text{He}(d,n)\) reaction (Q=17.59 MeV) at incident deuteron energies 1, 2, 3, and 4 MeV. Solid-state Ti/T target of 2 mg/cm\(^2\) thickness was used. The standard spread of the neutron energy distribution was calculated based of energy loss of the projectile in the target and the energy distribution of the neutrons for the sample irradiation geometry. The time profiles of the neutron source intensity during irradiations were recorded by long counter. The DT neutron spectrum from solid tritium target shows presence of low energy secondary neutrons at incident deuteron energies above 1 MeV. The neutron flux energy distribution for each irradiation was determined by time-of-flight spectrum measurement in combination with threshold activation monitor reactions measurements.

The studied reactions cross sections were calculated relative to the \(^{27}\text{An}(n,a)^{24}\text{Na}\) (ENDF/B-VI) standard cross section and \(^{93}\text{Nb}(n,2n)^{90}\text{Nb}\) reaction cross sections.

2.2. Measurement of radioactivity

The Standard gamma-ray spectrometry was employed for the measurement of the radioactivity induced in the samples during irradiation. The decay data used in the analysis are listed in Table 1.

The response of the HPGe detector for the measurement geometry was calculated by Monte Carlo simulations using the MCNP5 code [6] to account for the gamma-ray attenuation and the distribution of the induced activity in the large samples. The precise detector geometry, including the thickness of the dead layers on the outer surfaces of the crystal, was determined by comparisons of the calculated values with the measurements using calibration sources at various positions on the detector cap. The following gamma-ray standard sources, supplied by LEA Laboratoires Etalons d’Activité, France, were used for the calibration: \(^{241}\text{Am}\) (59.5412 keV), \(^{109}\text{Cd}\) (88.0336 keV), \(^{57}\text{Co}\) (122.0665 keV and 136.47256 keV), \(^{139}\text{Ce}\) (165.8575 keV), \(^{51}\text{Cr}\) (320.0824 keV), \(^{137}\text{Cs}\) (661.657 keV), \(^{54}\text{Mn}\) (834.848 keV), \(^{60}\text{Zn}\) (1115.539 keV), \(^{22}\text{Na}\) (1274.53 keV), and \(^{60}\text{Co}\) (1173.228 keV and 1332.492 keV). The uncertainty of the calculated efficiency was estimated by variation of the parameter values. For the variation the values of the parameters were randomly selected from the Normal distribution assuming 5 % standard deviation of the main value. Covariances between the uncertainties of the detector efficiencies for the studied reactions \(\varepsilon_r\) and for the monitor reaction \(\varepsilon_{st}\) were calculated as well. The uncertainty of the ratio \(\varepsilon_r/\varepsilon_{st}\) was determined by: var(\(\varepsilon_r/\varepsilon_{st}\)) = var(\(\varepsilon_r\)) + var(\(\varepsilon_{st}\)) - 2cov(\(\varepsilon_r,\varepsilon_{st}\)) [7].

The coincidence summing correction was applied in the case of \(^{59}\text{Fe}\) and \(^{24}\text{Na}\) activity measurement. The corrections were calculated based on the algorithm developed by Andreev at al. [8] using matrix formalism proposed in the work of Semkow at al. [9]. The uncertainty of the correction factor was calculated by propagation of the uncertainties of the peak and total efficiencies, \(\beta\) transition probability, \(\gamma\) transition + internal conversion probability and internal conversion coefficient [10].

Additional corrections applied in the data analysis were: neutron source intensity variation during irradiation, neutron flux and energy distribution inside the stack of the samples and monitor foils.

Due to the low cross sections for the multiple charged particle emission cross sections in the studied energy range the contribution from the interferences from reactions on other isotopes or
impurities in the sample were evaluated. The contribution from the $^{55}$Mn(n,2n)$^{54}$Mn was estimated at less than 1% based on the 100 ppm impurity in the sample and the assumption that the cross section is 800 mb. The estimated contribution from the $^{54}$Fe(n,p)$^{54}$Mn reaction is less than 0.1% based on 40 ppm concentration and 200 mb reaction cross section. The reactions $^{58}$Fe(n,$\gamma$)$^{59}$Fe, $^{59}$Co(n,p)$^{59}$Fe and $^{62}$Ni(n,$\alpha$)$^{59}$Fe on Fe, Co and Ni impurities in Cu sample are possible interfering reactions. However, activities from reactions with higher cross sections on these isotopes were not found in the measured spectra.

Table 1. Decay data of the measured reaction products ref. [11].

| Reaction                  | T_{1/2} (d) | E_γ (keV) | I_γ (%) |
|---------------------------|-------------|-----------|---------|
| $^{58}$Ni(n,p)$^{54}$Mn   | 312.19      | 834.848   | 99.9752 |
| $^{63}$Cu(n,p)$^{59}$Fe   | 44.949      | 1099.245  | 56.5131 |

3. Data analysis and uncertainty propagation

The cross sections were calculated using standard activation formula

$$\sigma_x = \sum \frac{A_x}{A_{st}} \frac{\varepsilon_x}{\varepsilon_{st}} \frac{t_{irr}}{t_{st}} \frac{f_{st}}{f_x} \frac{C_{x}^{\text{flux}}}{C_x^{st}} \frac{C_{x}^{\text{low}}}{C_x^{st}} \frac{C_{x}^{\text{coi}}}{C_x^{st}}$$

where $A$ is the number of the detected gamma rays in the full energy peak, $n$ is the number of the target nuclei, $\varepsilon$ is the detector efficiency, $f$ is the gamma-ray emission probability, $t_{irr}$ is the irradiation time, $t_c$ is the counting time, and measuring time $t_m$, $c_{x, st}^{\text{flux}}$ is the correction factor for neutron source intensity variation during irradiation, $c_{x, st}^{\text{low}}$ is the correction factor for the activity induced by low energy background neutrons, $c_{x, st}^{\text{coi}}$ is the gamma-rays true coincidence summing correction factor.

The standard uncertainties of the obtained cross sections were determined by propagation of the partial uncertainties of all parameters included in Eq. (1). The values of the partial uncertainties are given in tables 2 and 3 for $^{58}$Ni(n,p)$^{54}$Mn and $^{63}$Cu(n,p)$^{59}$Fe reaction, respectively. The uncertainties of the gamma-ray emission probabilities, the timing factors, samples’ mass, isotopic abundances and the uncertainties with estimated values, such as uncertainties of the reproducibility of the sample irradiation and counting geometry are combined in “others”.

Table 2. Partial and total uncertainties (%) of the $^{58}$Ni(n,p)$^{54}$Mn reaction cross section.

| E_n (MeV) | A_{Ni} | A_{Al} | $\varepsilon_{Al}/\varepsilon_{Ni}$ | $C_{Al}^{\text{low}}$ | $C_{Al}^{\text{coi}}$ | $C_{st}^{\text{flux}}$ | $C_{st}^{\text{low}}$ | $C_{st}^{\text{coi}}$ | $\sigma_{st}$ | others | total |
|-----------|--------|--------|-------------------------------|-----------------|-----------------|-------------------|-----------------|-----------------|-------------|--------|-------|
| 15.6      | 1.1    | 1.2    | 3.1                           | 0               | 0.4             | 1.6               | 2               | 5.9             | 1.1         | 1.2    | 3.1   |
| 16.8      | 2.1    | 2.2    | 3.1                           | 1.5             | 0.4             | 2.1               | 2               | 5.9             | 2.1         | 2.2    | 3.1   |
| 18.0      | 1.9    | 1.5    | 3.1                           | 1.5             | 0.4             | 2.2               | 2               | 5.6             | 1.9         | 1.5    | 3.1   |
| 19.1      | 1.4    | 2.0    | 3.1                           | 1.5             | 0.4             | 3.0               | 2               | 6.0             | 1.4         | 2.0    | 3.1   |
| 20.6      | 1.1    | 1.3    | 3.1                           | 1.5             | 0.4             | 5.1               | 2               | 7.0             | 1.1         | 1.3    | 3.1   |

Table 3. Partial and total uncertainties (%) of the $^{63}$Cu(n,p)$^{59}$Fe reaction cross sections.

| E_n (MeV) | A_{Cu} | A_{Al} | $\varepsilon_{Al}/\varepsilon_{Cu}$ | $C_{Al}^{\text{low}}$ | $C_{Al}^{\text{coi}}$ | $C_{st}^{\text{flux}}$ | $C_{st}^{\text{low}}$ | $C_{st}^{\text{coi}}$ | $\sigma_{st}$ | others | total |
|-----------|--------|--------|-------------------------------|-----------------|-----------------|-------------------|-----------------|-----------------|-------------|--------|-------|
| 17.4      | 3.5    | 1.2    | 2.5                           | 0.5             | 0.4             | 1.6               | 2               | 5.9             | 3.5         | 1.2    | 2.5   |
| 19.0      | 1.7    | 1.0    | 2.5                           | 0.5             | 0.4             | 2.8               | 2               | 5.6             | 1.7         | 1.0    | 2.5   |
| 19.9      | 1.7    | 2.7    | 2.5                           | 1.5             | 0.4             | 4.5               | 2               | 6.0             | 1.7         | 2.7    | 2.5   |
4. Results and discussion
The cross sections obtained in this work are presented in table 4 and table 5. The preliminary results [12] were finalized applying improved calculations of the coincidence summing correction factors and uncertainty analysis. The results are compared with the experimental data from EXFOR and evaluated nuclear data on figures 1 and 2.

![Figure 1](image1.png)  
**Figure 1.** Results for the $^{58}\text{Ni}(n,p\alpha)^{54}\text{Mn}$ reaction cross sections compared with the evaluated nuclear data.

![Figure 2](image2.png)  
**Figure 2.** Results for the $^{63}\text{Cu}(n,p\alpha)^{59}\text{Fe}$ reaction cross sections compared with the data from EXFOR and evaluated nuclear data.

There are good agreement with the Iwasaki data from 1993 [13] for the $^{58}\text{Ni}(n,p\alpha)^{54}\text{Mn}$ reaction cross section. However, the data from 1994 tend to be higher. The best agreement is found with the JEFF-3.3 evaluation.

There are no other experimental data for the $^{63}\text{Cu}(n,p\alpha)^{59}\text{Fe}$ cross section in EXFOR database. There are good agreements with EAF-2010 and JENDL/AD-2017 evaluations.

**Table 4.** Measured reaction cross sections for the $^{58}\text{Ni}(n,p\alpha)^{54}\text{Mn}$ reaction the cross section. Standard uncertainties are given for the cross sections. The standards spreads are given for the neutron energy distribution.

| $\text{En (MeV)}$ | $\sigma (\text{mb})$ |
|------------------|---------------------|
| 15.6 ± 0.3       | 2.97 ± 0.2          |
| 16.8 ± 0.3       | 7.06 ± 0.37         |
| 18.0 ± 0.2       | 17.4 ± 2.0          |
| 19.1 ± 0.2       | 26.2 ± 1.4          |
| 20.6 ± 0.1       | 35.4 ± 3.0          |
Table 5. Measured reaction cross sections for $^{63}$Cu(n,p)$^{59}$Fe reaction cross section. Standard uncertainties are given for the cross sections. The standards spreads are given for the neutron energy distribution.

| $E_n$ (MeV) | $\sigma$ (mb) |
|-------------|---------------|
| 17.4 ± 0.2  | 0.0453 ± 0.0026 |
| 19.0 ± 0.2  | 0.196 ± 0.013  |
| 19.9 ± 0.2  | 0.362 ± 0.03   |

5. Summary
This paper presents the results from the $^{58}$Ni(n,p)$^{54}$Mna and $^{63}$Cu(n,p)$^{59}$Fe activation cross section measurements in the energy range from 14 to 21 MeV. The induced activities were determined by gamma ray spectrometry. The cross sections were determined relative to $^{27}$Al(n,$\alpha$)$^{24}$Na standard cross section. The data for the $^{63}$Cu(n,p)$^{59}$Fe reaction was obtained for the first time.

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