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Newly developed semi-empirical formulas for (p, α) at 17.9 MeV and (p, np) at 22.3 MeV reaction cross-sections

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Abstract. In this study, we have investigated the asymmetry term effect for the (p, α) and (p, np) reaction cross-sections and obtained new coefficients for the (p, α) and (p, np) reaction cross-sections at 17.9 and 22.3 MeV energies. We have suggested semi-empirical formulas including the non-elastic effects of optical model found by fitting two parameters for proton-induced reactions. The coefficients were determined by least-square fitting method. The obtained cross-section formulas with new coefficients have been discussed and compared with the available experimental data.

Keywords. (p, α) and (p, np) reaction cross-sections; empirical formulas; optical model; pairing effect; asymmetry term effect.

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1. Introduction

The intermediate-energy proton-induced reactions can be used to produce radionuclides and these radionuclides can be used in medicine and industry. The high-energy proton-induced reactions are also required for advanced nuclear systems, such as spallation reactions for the production of neutrons in spallation neutron source (capable of incinerating nuclear waste and of producing energy), high-energy proton-induced fission as an alternative for the isotope production etc. [1,2]. Recently, many experimental techniques have been developed to measure the cross-sections of different particle-induced reactions. Nowadays, the experimental cross-sections are available in EXFOR/CSISRS data files as well as in the evaluated data files (such as ENDF/B-VII, JENDL-HE and JEEF-3.1) [3,4]. These data files contain information for a few tens of stable nuclides. But, still this is not enough for a complete and detailed study of activation and transmutation of materials irradiated with intermediate-energy protons [4–6]. To satisfy the growing needs of nuclear data at intermediate energies, the proton activation data file (PADF) has been prepared [7–9].

The evaluated nuclear cross-section data by producing radionuclides are required for understanding the binding energy systematics, nuclear structure, the properties of the excited nuclear states in different energy ranges and refined nuclear models [3–5]. These data are also necessary to develop more nuclear reaction models in order to explain nuclear reaction mechanisms and the properties of the excited nuclear states in different energy ranges. These nuclear reaction models are frequently needed to provide estimates of the particle-induced reaction cross-sections, especially if the experimental data are not available, or when it is hopelessly impractical to measure the cross-sections due to the experimental difficulty [6–9]. So the nuclear reaction systematics is widely used for the evaluation of reaction cross-section to supplement the result of measurements and calculations by theoretical models.

2. Empirical and semi-empirical systematics for the fast neutron-induced reactions

A large number of empirical and semi-empirical formulas with different parameters for cross-section calculations have also been proposed by several authors [10–12]. The applications of statistical and thermodynamical methods to calculate the nuclear process go back to the fundamental work of Weisskopf [13]. The empirical formulas have been proposed by Levkovskii for predicting (n, p) and (n, α) cross-sections at 14–15 MeV [14]. These formulas can be given as follows:

$$\sigma_{n,p} \approx \sigma_n \exp[-33(N-Z)/A], \quad \sigma_{n,\alpha} \approx \sigma_\alpha \exp[-33(N-Z)/A], \qquad (1)$$

where

$$\sigma_n = \pi r_0^2 (A^{1/3}+1)^2 \quad \text{and} \quad \sigma_\alpha = 0.4\pi r_0^2 (A^{1/3}+1)^2$$

with $r_0 = 1.2 \times 10^{-13}$ cm. It was shown that the formulas of Levkovskii were roughly equivalent to the theoretical approximations for 14–15 MeV neutron cross-sections derived on the basis of the effective Q-value. The (n, p) experimental cross-sections decrease smoothly with increasing mass number depending on the Q-value of a given element [15]. But the (n, α) experimental cross-sections-to-theoretical cross-sections ratio was found appreciably closer to unity when derived using the average Q-value, $Q_{\mathrm{av}} = (Q_{\mathrm{t}} + Q_{\mathrm{e}})/2$, where Q_{t} and Q_{e} are respectively the true and effective Q-values. This makes it difficult to discuss the theoretical relation between the empirical formulas and effective Q-values in the case of (n, α) cross-sections. According to previous reports, the cross-sections for many nuclei significantly vary with the mass number A, neutron number N and proton number Z of the target nucleus [10–16]. In addition, the effects attributable to the asymmetry parameter,

$s = (N - Z)/A$, as well as to the isotopic, isotonic and odd–even properties of the nuclei have been observed in the data. From the earlier works, the empirical cross-section of the reactions induced by fast neutrons can be approximately expressed as follows:

$$\sigma(n, x) = C\sigma_{\rm ne}\exp(as), \tag{2}$$

where x represents the particle produced by the reaction, $\sigma_{\rm ne}$ is the neutron non-elastic cross-section ($\sigma_{\rm ne} = \pi r_0^2(A^{1/3} + 1)^2$) and the coefficients C and a are the fitting parameters determined from least-squares method for different reactions. The fast neutron non-elastic cross-section is given by πR^2, where R is the nuclear radius. The exponential term represents the escape of the reaction products from a compound nucleus.

The pre-equilibrium processes are important mechanisms in nuclear reactions induced by light projectiles with incident energies above 10 MeV [17,18]. The pre-equilibrium reaction effects strongly depend on the asymmetry parameter. The researches have shown that the fast neutron-induced reaction is possible with equilibrium (compound) process in region I (A = 40–62). But, in region III (A = 90–160), this reaction is possible with the pre-equilibrium process and in the intermediate region II (A = 63–89), the reaction is governed by both equilibrium and pre-equilibrium processes [15,19,20]. Kumabe and Fukuda have modified the Levkovskii's formula by taking this into consideration for (n, p) and (n, α) cross-sections at 14–15 MeV [15]. This formula is given as follows:

$$\sigma(n, x) = aA^b\exp[-c\,(N - Z)/A], \tag{3}$$

where x represents p or α, and separately for each of the above three regions, the coefficients a, b and c are determined by a least-squares fitting. Equation (3) can be in agreement with the experimental data if A^b ($b > 2/3$) is replaced by $(A^{1/3} + 1)^2$. But it ignores the physical meaning of the neutron non-elastic cross-section σ_n and σ_α.

More recently, new empirical formulas have been developed including the neutron non-elastic cross-section $\sigma_{\rm ne}$ for 14–15 MeV neutron-induced reaction cross-sections. Tel *et al* suggested using these new experimental data to develop a new empirical formula for the cross-sections of the (n, p) reactions [19]. This formula depends on the asymmetry parameter, $s = (N - Z)/A$, and has the pairing effect on the binding energy systematics of nuclear shell model. Tel *et al* also obtained a new appropriate coefficient by using this formula for $(n, 2n)$, (n, α), (n, t) and (n, d) reactions [19–24]. Betak *et al* have applied this formula for (n, p) reactions and have found agreement with odd-A nucleus in the latest experiment for $^{112-124}$Sn thin target [25]. Although the parameters obtained by Tel *et al* provide a means for fitting a number of such cross-sections at 14–15 MeV, there are some cross-sections which deviate substantially from the obtained fit results. In the latest work, Hadizadeh and Grimes examine straightforward extensions of the model to see if the fit can be improved [26].

The cross-section evaluation for materials irradiated by protons attends special importance to use of systematic of proton-induced reaction cross-section. Although there are a large number of cross-section formulas for the neutron-induced reactions, the number of proton-induced reactions, for which the systematic dependence of cross-section can be investigated using experimental data, is rather small.

The first systematics for (p, n) reaction cross-sections were discussed by Broeders and Konobeyev [7,27]. The first systematics for (p, xn) reactions have been obtained by Röhm *et al* [28] and Münzel *et al* [29]. The semi-empirical formulas for the (p, α), $(p, n\alpha)$ and (p, np) reaction cross-sections were derived by using analytical expressions describing the equilibrium and non-equilibrium particle emissions in nuclear reactions by Broeders and Konobeyev [7]. Their work for semi-empirical systematics of (p, α) and (p, np) reaction cross-sections were obtained at 17.9 and 22.3 MeV incident proton energies [7]. The semi-empirical systematics are based on analytical formulas derived from the pre-equilibrium exciton model, evaporation model and semi-empirical mass formula. Finally, Tel *et al* have obtained a new appropriate coefficient for the $(p, n\alpha)$ reactions at 24.8 and 28.5 MeV energies [30].

The best fitting can be obtained with the new free parameters in order to provide the minimum value of the following expression:

$$\chi^2 = \frac{1}{N} \sum_{i}^{N} \left(\frac{\sigma^i_{\rm exp} - \sigma^i_{\rm cal}}{\Delta\sigma^i_{\rm exp}} \right)^2, \tag{4}$$

where $\sigma^i_{\rm exp}$ and $\sigma^i_{\rm cal}$ are the experimental and the calculated cross-sections, respectively, and $\Delta\sigma^i_{\rm exp}$ is the error associated with $\sigma^i_{\rm exp}$. For the empirical and semi-empirical formulas, details on the results of the best fitting parameters, the values of χ^2 and correlation coefficient R^2 can be found for (n, charged particle), $(n, 2n)$, (p, α) and (p, np) reactions in refs [7,10–12,19–24,30].

3. Results and discussion

The researches have shown that the (n, p) and (n, α) cross-sections are strongly dependent on their Q-values [31,32]. If the Q-value decreases smoothly with increasing mass number for a given element, the cross-sections also decrease smoothly. The actual experimental (n, p) and (n, α) cross-sections decrease smoothly with increasing mass number for a given element [15], despite the large differences in true Q-value between magic and non-magic nuclei, and between even-Z-even-N and odd nuclei. This behaviour can be explained in terms of the effective Q-value [31–35]. The resulting Q-value effects have a strong $(N - Z)/A$ dependence implied by eq. (2). And also some works were done to analyse the shapes of excitation functions and cross-sections at its maximum [33]. Konno *et al* suggested using the empirical formula of the (n, np) reactions including neutron non-elastic cross-section [35];

$$\begin{aligned} \sigma_{n,np} &= 68.46(A^{1/3}+1)^2 \exp(-25.08s), \quad \text{for } Sn > Sp, \\ \sigma_{n,np} &= 22.05(A^{1/3}+1)^2 \exp(-31.91s), \quad \text{for } Sn < Sp, \end{aligned} \tag{5}$$

where Sn and Sp are neutron and proton separation energies, respectively.

In this study, the same relation can be expected for the (p, α) and the (p, np) reaction cross-sections. In this respect, the proton energies, vicinities of which contain the maximal number of cross-section measurements for each reaction at a wide range of target nuclei, have been taken. We have selected energy points for

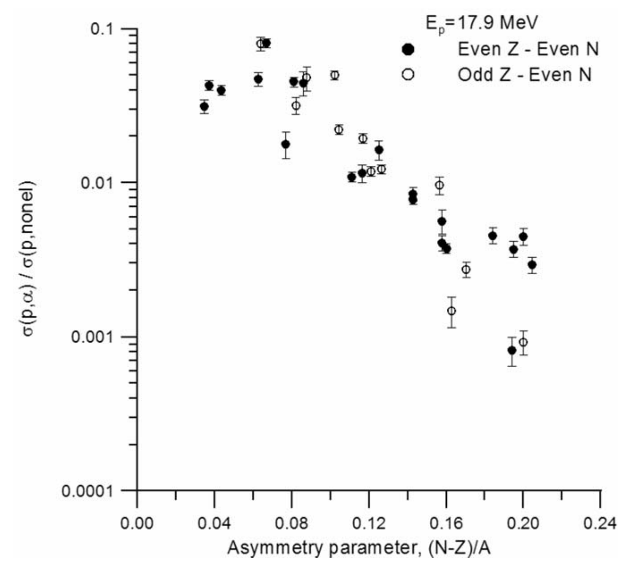

Figure 1. Systematics of (p, α) reactions (in mb) at 17.9 MeV protons. Experimental values were taken from refs [3,7].

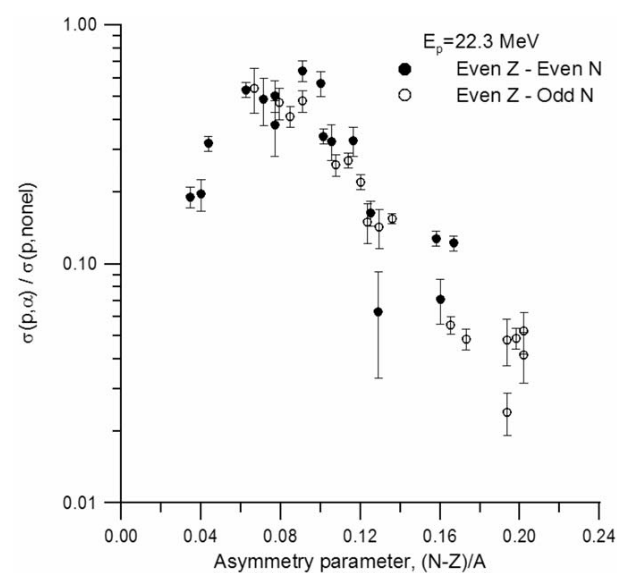

Figure 2. Systematics of (p, np) reactions (in mb) at 22.3 MeV protons. Experimental values were taken from refs [3,7].

determining the cross-sections at the maximum of the reaction excitation functions. It has been shown that the method of 'statistical analysis of cross-sections' (SACS) can be used for this purpose. This work is similar in spirit to that of Forrest and Kopecky [36]. These selected energy points are at 17.9 MeV for the (p, α) reaction and at 22.3 MeV for the (p, np) reaction.

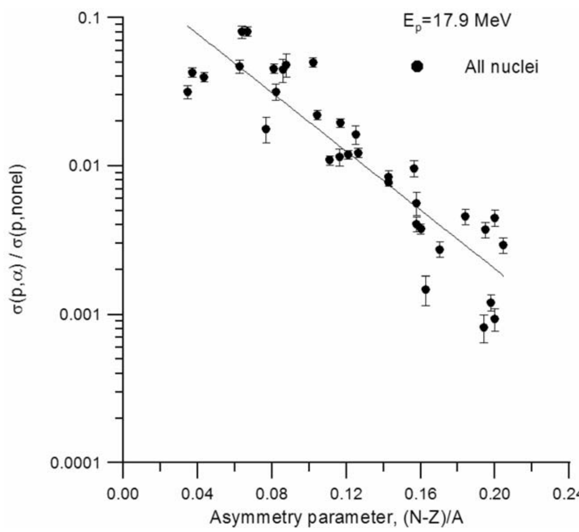

Figure 3. Systematics of (p, α) reactions (in mb) at 17.9 MeV for all nuclides used in this work. Experimental points were fitted with eq. (6). The obtained coefficients C, a and R^2 are given in table 1.

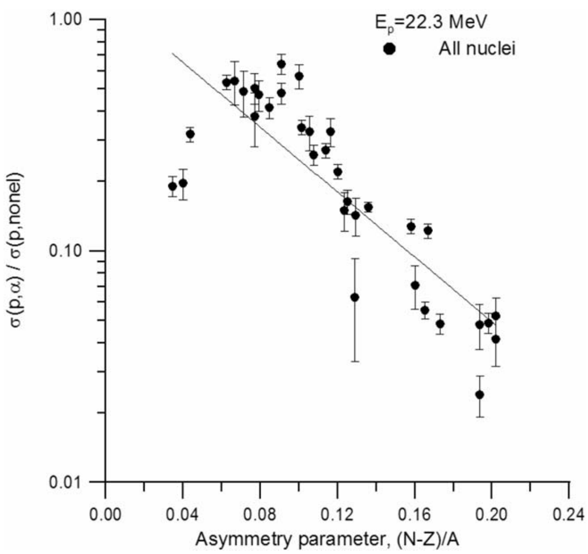

Figure 4. Systematics of (p, np) reactions (in mb) at 22.3 MeV for all nuclides used in this work. Experimental points were fitted with eq. (6). The obtained coefficients C, a and R^2 are given in table 2.

The measured experimental (p, np) reaction cross-sections were taken from the activation method and contain contributions of (p, np) and (p, d) reaction cross-sections. As can be obviously seen from figures 1 and 2, the measured (p, α) and (p, np) reaction cross-sections exhibit a gradient (increases for the lighter mass nuclei) with increasing asymmetry parameter, and then become maximum (for

Table 1. The obtained coefficients C and a, and the empirical formulas for (p,α) reactions at 17.9 MeV.

Z	N	C	a	$\sigma(p,\alpha)/\sigma_{(p,\,\text{nonel.})} = C\exp(as)$	R^2
All nuclei		0.1930	−22.7961	$\sigma(p,\alpha)/\sigma_{(p,\,\text{nonel.})} = 0.1930\exp(-22.7961s)$	0.82
Even	Even	0.1211	−19.2372	$\sigma(p,\alpha)/\sigma_{(p,\,\text{nonel.})} = 0.1211\exp(-19.2372s)$	0.85
Even	Odd	0.7891	−33.3204	$\sigma(p,\alpha)/\sigma_{(p,\,\text{nonel.})} = 0.7891\exp(-33.3204s)$	0.92

Table 2. The obtained coefficients C and a, and the empirical formulas for (p,np) reactions at 22.3 MeV.

Z	N	C	a	$\sigma(p,np)/\sigma_{(p,\,\text{nonel.})} = C\exp(as)$	R^2
All nuclei		1.2413	−16.1266	$\sigma(p,np)/\sigma_{(p,\,\text{nonel.})} = 1.2413\exp(-16.1266s)$	0.70
Even	Even	0.6667	−9.7733	$\sigma(p,np)/\sigma_{(p,\,\text{nonel.})} = 0.6667\exp(-9.7733s)$	0.32
Odd	Even	2.4871	−21.0790	$\sigma(p,np)/\sigma_{(p,\,\text{nonel.})} = 2.4871\exp(-21.0790s)$	0.96

medium heavy mass nuclei) and decrease with increasing asymmetry parameter (for heavier mass nuclei).

We assumed that the semi-empirical cross-sections of reactions induced by proton can be approximately expressed as follows:

$$\sigma(p,x)/\sigma_{(p,\,\text{nonel.})} = C\exp(as), \tag{6}$$

where x represents particles of the reaction produced. The coefficients C and a are the fitting parameters determined from the least-squares method. The $\sigma_{p,\text{nonel.}}$ represents the proton non-elastic cross-section. In this work, the $\sigma_{p,\text{nonel.}}$ proton non-elastic cross-sections were obtained by using the optical potential parameters of Becchetti and Greenlees [38]. The proton non-elastic cross-section calculations have been done in the framework of the optical model using SCAT 2 computer code [39,40].

For fitting procedure, we have used 34 experimental data taken from refs [3,7] with their mass numbers A from 46 to 197 and atomic numbers Z from 22 to 79, for the (p,α) reaction cross-sections at 17.9 MeV. And also, we have used 34 experimental data taken from refs [3,7] with their mass numbers A from 45 to 203 and atomic numbers Z from 21 to 81 for the (p,np) reaction cross-sections at 22.3 MeV.

Firstly, the fitting parameters of empirical formula have been determined for all target nuclei having mass number for (p,α) and (p,np) reaction cross-sections at 17.9 and 22.3 MeV (in figures 3, 4 and in tables 1, 2). Secondly, we have determined two different fitting parameter groups by classifying the target nuclei into even–even and odd–even (in figures 5–8 and in tables 1, 2). The comparison of the first fitting and the second fitting reaction cross-section formulas shows that the second formula proposed in our study gives better description of experimental data. From tables 1 and 2, it can be seen that there is a better correlation because R^2 is higher than

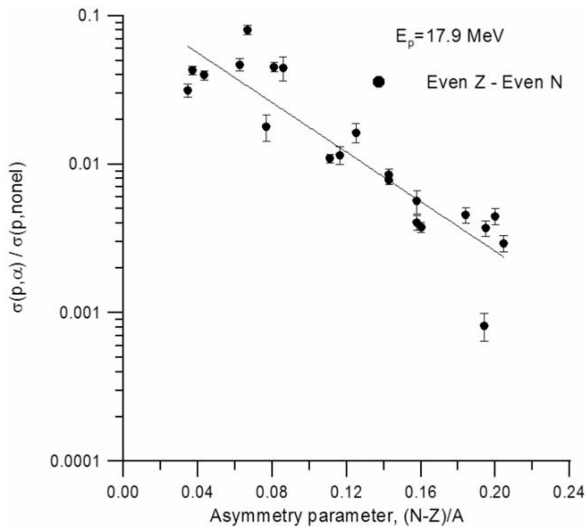

Figure 5. Systematics of (p, α) reactions (in mb) at 17.9 MeV for even-Z, even-N nuclides. Experimental points were fitted with eq. (6). The obtained coefficients C, a and R^2 are given in table 1.

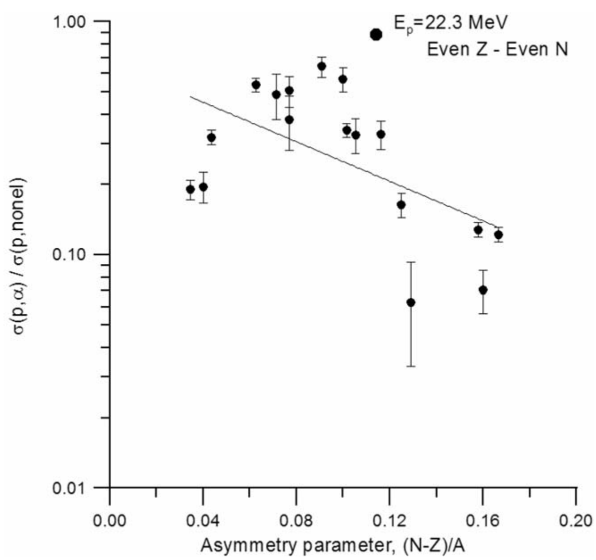

Figure 6. Systematics of (p, np) reactions (in mb) at 22.3 MeV for even-Z, even-N nuclides. Experimental points are fitted with eq. (6). The obtained coefficients C, a and R^2 are given in table 2.

the first fitting procedure. The maximum R^2 values of the second formula provide closer results to the experimental data. Figures 9–12 compare the ratios of the experimental (p, α) and (p, np) cross-sections to those calculated using formulas given in tables 1 and 2. These show an improvement in describing the (p, α) and

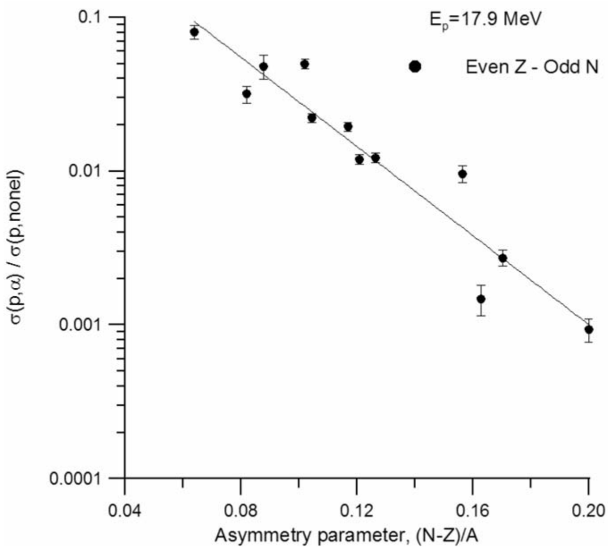

Figure 7. Systematics of (p, α) reactions (in mb) at 17.9 MeV for even-Z, odd-N nuclides. Experimental points were fitted with eq. (6). The obtained coefficients C, a and R^2 are given in table 1.

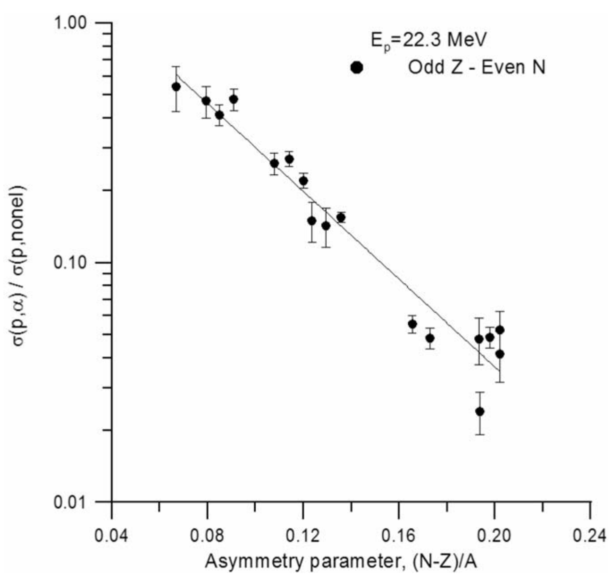

Figure 8. Systematics of (p, np) reactions (in mb) at 22.3 MeV for odd-Z, even-N nuclides used in this work. Experimental points are fitted with eq. (6). The obtained coefficients C, a and R^2 are given in table 2.

(p, np) reaction cross-sections compared with the first formula. The second formula includes the odd–even (and even–even) nuclei correction and this formula has been taken into consideration to be the pairing effects.

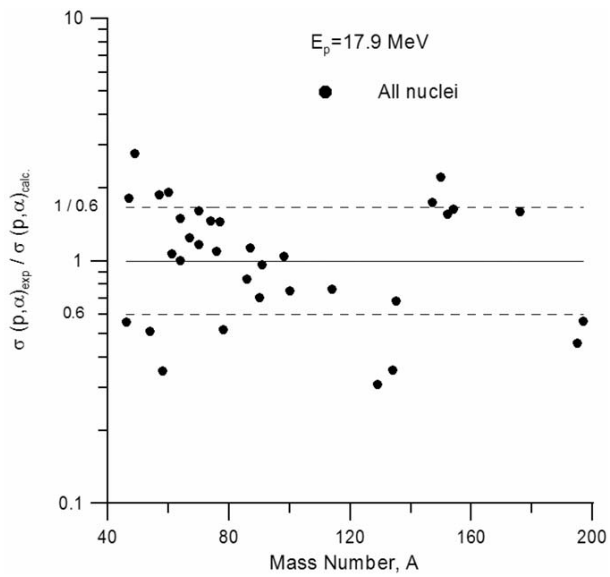

Figure 9. Ratios of the experimental to the calculated (p,α) cross-sections at 17.9 MeV using formulas given in table 1.

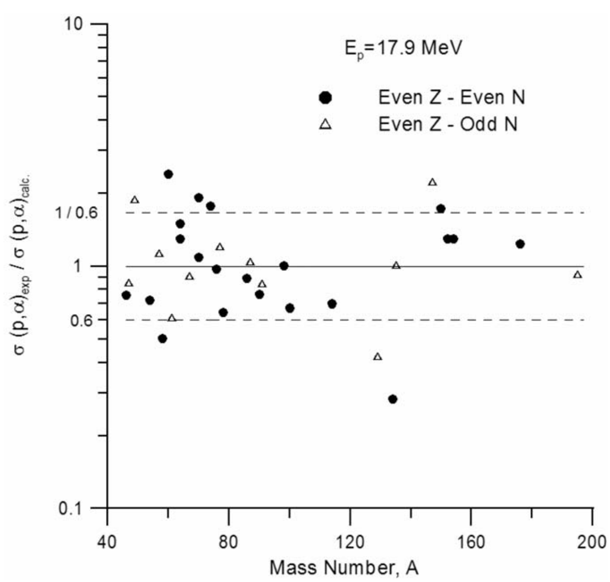

Figure 10. Ratios of the experimental to the calculated (p,α) cross-sections at 17.9 MeV using even–even and even–odd nuclides fitting formula given in table 1.

The semi-empirical cross-section formula including proton optical model non-elastic cross-section with pairing interaction is the best fitting formula for proton-induced reaction. These semi-empirical formulas use the evaporation model

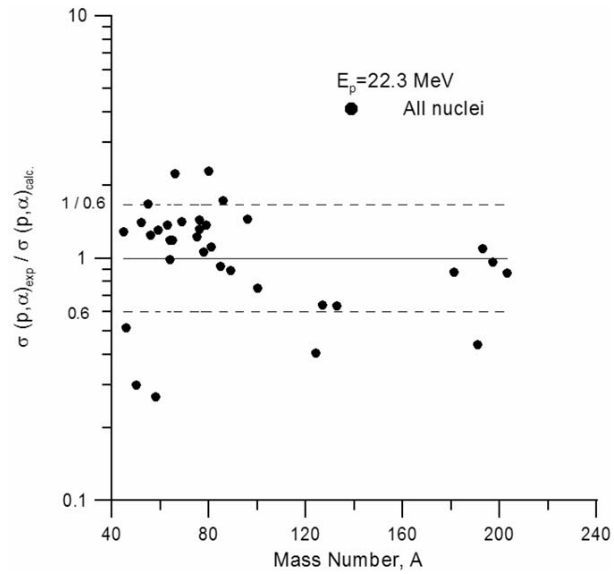

Figure 11. Ratios of the experimental to the calculated (p, np) reactions at 22.3 MeV using the nuclides fitting formula given in table 2.

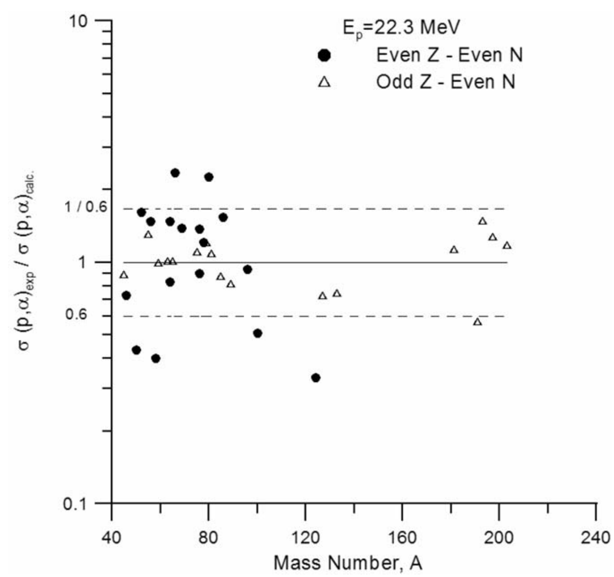

Figure 12. Ratios of the experimental to the calculated (p, np) reactions at 22.3 MeV using even–even and even–odd nuclides fitting formula given in table 2.

and ignore the importance of the pre-equilibrium mechanism of particle emission for medium and heavy nuclei. The semi-empirical formulas including pre-equilibrium mechanism are in better agreement than the present empirical formulas in this work because the other semi-empirical systematics are based on analytical expressions for the calculation of particle emission within the frame of pre-equilibrium exciton and evaporation models. The present formulas given in tables 1 and 2 for the (p, α) and (p, np) reactions can still be considered as a very useful practical tool for estimating quickly and with relatively good accuracy the cross-sections in question, in view of the poor agreement with experiment sometimes provided by calculations based on the semi-empirical systematics. If more experimental data can be obtained using the new technology (especially, even–even, even–odd, odd–even and odd–odd target nuclei) for the (p, α) and (p, np) cross-sections, semi-empirical systematics, nuclear reaction mechanisms and nuclear models could be developed in the desired way. As a result, the present studies lead to improve and clarify the binding energy systematic of the nuclear shell model.

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