### ЯДЕРНА ФІЗИКА NUCLEAR PHYSICS

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## EVALUATED EMPIRICAL SYSTEMATICS OF $(n, n\alpha)$ REACTION CROSS-SECTIONS AT 14 - 15 MeV INCIDENT ENERGY

In this study, the asymmetry parameter-included empirical equation proposed by Tel et al. has been applied for  $(n, n\alpha)$  reaction at 14 - 15 MeV energies. The free parameters of equation have been determined by fitting of experimental cross-section data. Even-even and odd-even nuclei classification have been made in order to taking into account of pairing effect on the  $(n, n\alpha)$  reaction cross-sections. Excitation function calculations have been carried out for target nuclei such as  $^{65}$ Cu,  $^{71}$ Ga,  $^{31}$ P and  $^{99}$ Tc by  $(n, n\alpha)$  reactions up to 30 MeV neutron induced energy. Comparisons have been made among available experimental data and results of cross-sections calculated by nuclear models with results of values calculated by empirical formulas. Determined relations are functional for the  $(n, n\alpha)$  reaction cross-sections at 14 - 15 MeV incoming neutron energies. Acceptable harmonies have been seen between collected experimental data and modelled calculations.

*Keywords:* empirical formulas, asymmetry term,  $(n, n\alpha)$  reactions, cross-section systematics, nuclear reaction codes.

#### 1. Introduction

Cross-section values of neutron-induced reactions in the energy range below 20 MeV are rather precious for fast neutron included applications such as fission and accelerator-driven reactor systems. Neutron induced reaction cross-sections are of fundamental importance for neutron transport, neutron regeneration technologies. Also, better valued accurate cross-section data is needed for the nuclear and shielding design of fission reactors and accelerator-based systems [1, 2]. Elastic-inelastic scattering and radiative capture reactions are possible at 14 - 15 MeV energies. Many complex and multiple particle emission reaction channels such as (n, 2n), (n, p),  $(n, \alpha)$ , (n, np), (n, d), (n, 3n) and  $(n, n\alpha)$  are opened at this energy range. Cross-section variations and spectrums have a great sign for excited nuclei and nuclear structure. The data is required to understand the nuclear reaction mechanism and to develop and test the validity of available nuclear reaction models. Because of the certain problems such as experimental handicaps and poor economic conditions, nuclear models have been utilized to obtain neutron-induced reaction cross-sections for a long time [3 - 7]. Reliable experimental data is important for comparing with theoretical nuclear model calculations for testing predictive ability.

In the present work, we have performed parameterrization study based on simple empirical relation which contains free parameters, in order to estimate cross-section values for  $(n, n\alpha)$  reaction about 14-15 MeV energies. Even-even and odd-even nuclei categorizations have been carried out. Results of crosssection calculations are plotted for the following targets with neutron-induced reactions: <sup>65</sup>Cu, <sup>71</sup>Ga, <sup>31</sup>P and <sup>99</sup>Tc. The modeled excitation function calculations have been carried out by using ALICE/ASH, Empire 3.2 and Talys 1.6 codes in the framework of equilibrium and pre-equilibrium nuclear reaction models. The obtained results have been discussed and compared with the available experimental data and the calculated empirical values. As a result, a good agreement was found between our results and experimental data.

# 2. Empirical systematic for 14 - 15 MeV $(n, n\alpha)$ reaction cross-sections

In general, nuclear reaction models based on statistical and thermodynamic approach consist of more complex formulas. However, empirical and semi-empirical reaction cross-section studies suggest easily understandable equations containing free parameters at the end of analyzing the experimental data [8 - 13]. It is known that there is a strong dependence with the reaction cross-section and the target nuclei mass number *A*, neutron number *N*, proton number *Z*. According to previous studies [14 - 19], this dependence explains the isotonic, isotopic property of target nuclei.

The empirical cross-sections of reactions induced by fast neutrons can be approximately expressed as follows

$$\sigma(n,x) = C\sigma_{ne} exp[as], \qquad (1)$$

$$\sigma_{ne} = \pi r_0^2 \left( A^{1/3} + 1 \right)^2,$$
 (2)

where  $\sigma_{ne} = \pi R^2$  (where *R* is the nuclear radius) sym-

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bolize target nuclei mass number dependent neutron non-elastic cross-section and  $r_0 = 1.2 \cdot 10^{-13}$  cm is the effective radius constant, s = (N - Z)/A is the asymmetry parameter. The coefficients C and a are the fitting parameters determined from least squares method for different reactions. Each one of Eq. (1) terms matches with nuclear reaction mechanism stages. The separation of the reaction particles from a compound nucleus is indicated by the exponential factor. Several empirical formulas were suggested to exhibit relationship between cross-sections with target nuclei numbers for the neutron-induced reactions at 14 - 15 MeV. Levkovsky proposed an empirical relation that describes the isotopic dependence of the (n, p) and  $(n, \alpha)$  cross-sections at an energy of 14.5 MeV [15, 16]. Levkovsky's relations were the foundation of following systematic empirical works. S. Qaim and G. Stocklin analyzed systematics of (n, t) reaction cross-section [20]. Belgaid and Asghar [21] and Ait-Tahar [22] studied (N-Z+1)/A term effects at the  $(n, \alpha)$  cross-sections. Q-binding energies and shell effects relations with the cross-sections were investigated for  $(n, \alpha)$  reaction at the 14 - 15 MeV energies [23, 24]. Also, in recent years many other empirical cross-section studies have focused on different nuclear reactions [25 - 29].

Neutron excess parameter or asymmetry parameter (N - Z)/A symbolize nuclear matter symmetry and neutron distribution in the nuclei. Asymmetry parameter value varies between 0 and 1; stable nuclei range up at 0.24. (N-Z)/A parameter significantly affects exponential term in the Eq. (1). The equation points out that, the probability of particle emission varies with the nuclear matter asymmetry (in other word proton and neutron number rate) [30]. (N - Z)/A dependence was studied for the  $(n, n\alpha)$  activation cross reaction and cross-section decrease as a function of (N - Z)/A term was appeared by Qaim [31]. Tel et al. [32] proposed a new empirical formula that depends on the asymmetry parameter for the (n, p) reaction cross-sections. Experimental data was evaluated, taking into account the pairing effects on the binding energy. Furthermore, this formula was applied and confirmed for (n, 2n), (p, n), (n, a), (n, d), and (n, t) reactions [33 - 38].

#### 3. Nuclear model calculations

ALCE/ASH [39], Empire 3.2 [40], and Talys 1.6 [41] are significant codes that are widely used by nuclear reaction researchers. The codes have been chosen in this study because of their calculation accuracy and simple utility. These nuclear codes involve several nuclear reaction models and too many parameters. It is possible that not only light nuclei, but also heavy nuclei calculations can be performed by means

of with these codes up to GeV energy scale. Equilibrium process at low energies is described with the Multiple Hauser - Flashbash evaporation theory in Talys and Empire codes. As for the ALICE/ASH code, Weisskopf - Ewing model is elected for the equilibrium emissions. Pre-equilibrium emissions were calculated using the Geometry Depended Hybrid model in ALICE/ASH. However, in Talys code, pre-equilibrium mechanism is explained according to the two-component exciton model and multiple pre-equilibrium emission method. Also, classical exciton model was chosen for the Empire 3.2. to calculate the pre-equilibrium process. Detailed information about these codes, nuclear models and parameters can be obtained from Refs. [39 - 41].

### 4. Results and discussions

In this study, asymmetry parameter dependent formula reported by Tel et al. [32] has been applied to neutron-induced  $(n, n\alpha)$  reaction and C and a free parameters have been determined. Proposed empirical and semi-empirical formulas include exponential relationship between the cross-section and the number of nucleons in the target nucleus.

The experimental  $(n, n\alpha)$  reaction cross-section systematics and the empirical fits have been plotted with nuclear asymmetry term (N - Z)/A in Fig. 1. Plots of  $\sigma/(A^{1/3}+1)^2$  and s demonstrate that the cross-section values decline as the asymmetry parameter increases. Charged particle emission rate has increased with light nuclei to heavy nuclei, i.e. with raising proton number [30]. Contribution of compound process emissions is shown as up to 10 - 15 MeV whereas pre-equilibrium

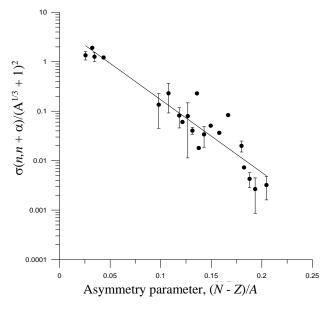


Fig. 1. The experimental points were fitted with  $\sigma_{empirical}$   $(n, n\alpha) = 5.03(A^{1/3} + 1)^2 exp(-33.87s)$  and correlation coefficient was determined as  $R^2 = 0.88$ . Experimental data were taken from the EXFOR [42] file.

mechanism has an essential role for the nuclear reactions above 10 - 15 MeV energies. The pre-equilibrium process is affected by asymmetry parameter (N - Z)/A. Investigations show that the compound nucleus mechanism governs the nuclear reaction process at the nuclei mass number range of  $A = 40 \sim 62$ , while the pre-compound mechanism affects at a range of  $A = 90 \sim 160$ . The reaction is dominated by the two processes when the target nuclei mass number valued between 63 and 89 [12].

In Fig. 2, the target nucleus cross-sections have been classified into two main group as even-Z, even-N nuclides and odd-Z, even-N nuclides. Expe-

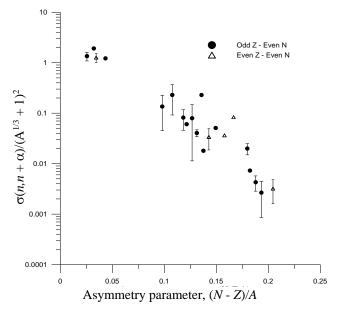


Fig. 2. Systematic of  $(n, n\alpha)$  reaction cross-sections (in mb) for odd-Z, even-N; even-Z, even-N nuclides induced by 14 - 15 MeV neutrons.

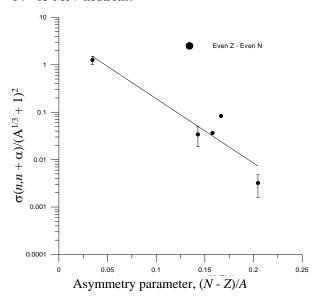


Fig. 4. Systematic of  $(n, n\alpha)$  reaction cross-sections (in mb) for even-Z, even-N nuclides induced by 14 - 15 MeV neutrons. Correlation coefficient was determined as  $R^2 = 0.86$ .

rimental odd-even nuclide cross-section systematics were grouped in Fig. 3 as well as even-even's in Fig. 4. (N - Z)/A-cross-section dependence has been seen in Figs. 2 - 4, that shapely cross-section gradient is declining with the symmetry parameter value of 0 to 0.2. It can be clearly inferred from Figs. 1 - 4 that the  $(n, n\alpha)$  reaction cross-sections have a decrease with the increasing of asymmetry parameter. This dependence is an observed result from the previous studies and has been confirmed for  $(n, n\alpha)$  reaction. Therefore, cross-section systematics have formed shapely gradient, acceptable free parameter values have been obtained via fitting process.

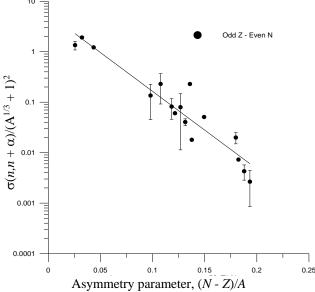


Fig. 3. Systematic of  $(n, n\alpha)$  reaction cross-sections (in mb) for odd-Z, even-N nuclides induced by 14 - 15 MeV neutrons. Correlation coefficient was determined as  $R^2 = 0.89$ .

Twenty-one collected (from EXFOR [42]) experimental  $(n, n\alpha)$  cross-section data for various target nuclei have been utilized for the fitting process. Five experimental cross-section data belong to even-even target nuclide, whereas sixteen experimental data belong to odd-even nuclides. The target nuclei mass number have changed between  $A = 23 \sim 181$ , proton number  $Z = 11 \sim 73$ , and neutron number  $N = 12 \sim 108$ . Empirical and semi-empirical cross-section parameterization have been carried out by using the plotted graphs in Figs. 1, 3, 4 via Tel et al. relation for  $(n, n\alpha)$ reaction about 14 - 15 MeV energy ranges. An empirical formula and two semi-empirical formulas that include pairing effect systematics have been formed along with three set of C and a parameters. Leastsquares method has been chosen for estimate these free parameters. Fitting reliability was ensured through the determined chi-square

$$\chi^{2} = \frac{1}{N} \sum_{i}^{N} \left( \frac{\sigma_{exp}^{i} - \sigma_{cal}^{i}}{\Delta \sigma_{exp}^{i}} \right)^{2}$$

correlation coefficient. Empirical and semi-empirical expressions for the  $(n, n\alpha)$  reactions about 14-15 MeV neutron energy, even-odd characteristics of formulas and  $\chi^2$  coefficient are given in Table 1.

In addition, cross-sections calculated by means of derived formulas and measured  $(n, n\alpha)$  reaction cross-sections are given in Table 2 and Fig. 5, respectively.

Table 1. The parameters C and a, and the empirical and semi-empirical formulas for  $(n, n\alpha)$  reactions

Z	N	C	а	$\sigma(n, n\alpha) = C\sigma_{ne} exp[as]$	$\chi^2$	
All Nuclei		5.03	-33.87	$\sigma_{emp}(n, n\alpha) = 5.03(A^{1/3} + 1)^2 exp[-33.87s]$		
Odd	Even	5.63	-35.36	$\sigma_{sem-emp}(n, n\alpha) = 5.63(A^{1/3} + 1)^2 exp[-35.36s]$	6.4	
Even	Even	4.43	-31.32	$\sigma_{sem-emp}(n, n\alpha) = 4.43(A^{1/3} + 1)^2 exp[-31.32s]$	4.3	

Table 2. The comparison of the cross-sections calculated with the empirical and semi-empirical formulas with the experimental (n, na) reaction cross-sections for 14 - 15 MeV incident neutrons.

Experimental data were taken from the EXFOR file

Target	Reaction products	Q-value, MeV	E <sub>excitation</sub> , MeV	$\sigma_{exp}$ , mb	$\Delta\sigma_{exp}, \ \mathrm{mb}$	σ <sub>empirical</sub> , mb	σ <sub>semi-empirical</sub> , mb
<sup>23</sup> Na	<sup>19</sup> F	-10.47	3.63	18	18	17.070	17.881
<sup>31</sup> P	<sup>27</sup> Al	-9.67	4.90	32.9	6.58	28.977	30.866
<sup>39</sup> K	<sup>35</sup> Cl	-7.22	6.88	26	5.1	40.764	38.277
51 <b>V</b>	<sup>47</sup> Sc	-10.29	3.81	3	2	4.034	3.896
<sup>58</sup> Ni	<sup>54</sup> Fe	-6.40	7.70	30	6	37.175	35.703
<sup>65</sup> Cu	<sup>61</sup> Co	-6.79	7.31	5.8	3.48	3.308	3.830
$^{70}$ Zn	<sup>66</sup> Ni	-5.96	8.74	0.89	0.4	1.046	0.944
<sup>71</sup> Ga	<sup>67</sup> Cu	-5.26	9.44	2.1	1.8	1.818	2.209
<sup>76</sup> Ge	<sup>72</sup> Zn	-7.51	7.19	1	0.2	0.657	0.864
$^{81}$ Br	<sup>77</sup> As	-6.48	8.22	6.5	1.3	1.437	1.787
<sup>87</sup> Rb	<sup>83</sup> Br	-8.01	6.69	1.5	0.3	0.942	1.213
<sup>96</sup> Zr	<sup>92</sup> Sr	-4.91	9.59	2.6	0.52	0.554	0.746
<sup>93</sup> Nb	<sup>89</sup> Y	-1.93	12.77	2.5	1.1	2.804	2.627
<sup>99</sup> Tc	<sup>95</sup> Nb	-2.97	11.73	1.28	0.2	1.866	1.715
<sup>107</sup> Ag	<sup>103</sup> Rh	-2.80	11.90	2	0.4	2.716	2.532
<sup>109</sup> Ag	<sup>105</sup> Rh	-3.29	11.41	0.6	0.12	1.589	1.447
<sup>139</sup> La	<sup>135</sup> Cs	-1.99	12.51	0.76	0.19	0.435	0.605
<sup>159</sup> Tb	<sup>155</sup> Eu	-0.14	14.56	0.3	0.6	0.430	0.366
<sup>165</sup> Ho	<sup>161</sup> Tb	0.14	14.84	0.18	0.06	0.365	0.308
<sup>176</sup> Yb	<sup>172</sup> Er	0.57	15.27	0.14	0.07	0.215	0.177
<sup>181</sup> Ta	<sup>177</sup> Lu	1.52	15.62	0.118	0.08	0.319	0.267

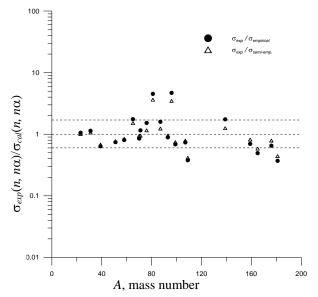


Fig. 5. Ratios of the  $(n, n\alpha)$  experimental cross-sections to the cross-sections calculated with empirical and semi-empirical formulas.

Above-mentioned nuclear reaction codes have been used to compare parameterized results to modelled calculations. The cross-sections for  $(n, n\alpha)$  reactions on  $^{65}$ Cu,  $^{71}$ Ga,  $^{31}$ P, and  $^{99}$ Tc targets in the energy range from threshold 30 MeV neutron incident energies have been estimated. Excitation function calculations have been obtained by the compound and pre-equilibrium nuclear reaction models for  $(n, n\alpha)$  reactions. Along with experimental data, graphics contain calculated empirical cross-section values at 14 - 15 MeV neutron-induced energy to enable comparisons to be made with model calculations.

The calculations with ALICE/ASH nuclear code modelled by the Weisskopf - Ewing theory and geometry depended hybrid model are generally in agreement with the measured cross-sections and the empirical calculations at the energy range of 10 - 20 MeV in Figs. 6 - 9. Moreover, these models are nearly in harmony with the other code calculations.

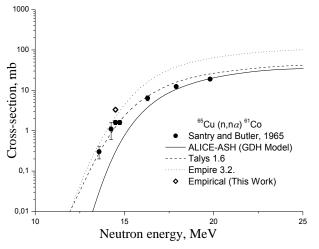


Fig. 6. The comparison of calculated excitation function of  $^{65}$ Cu $(n, n\alpha)^{61}$ Co reaction with the values reported in Ref. [42].

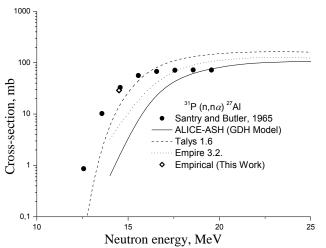


Fig. 8. The comparison of calculated excitation function of  $^{31}$ P(n,  $n\alpha$ ) $^{27}$ Al reaction with the values reported in Ref. [42].

Likewise, model-calculated results are compatible with the empirical-calculated value for target nuclei. The calculated cross-section results for  $(n, n\alpha)$  reaction are mainly from the contribution of the equilibrium emission process at these incident neutron energies.

Talys 1.6 code calculations for the equilibrium process with the Multiple Hauser - Feshbach decay model show good agreement with the experimental cross-sections for all reactions. As for the excitation functions in figures, equilibrium and pre-equilibrium components have not been demonstrated separately but together. These calculations include minimal pre-equilibrium effects at about 15 MeV. Within this energy range, the total compound cross-section, i.e., summed over all final discrete states and the excited continuum, is however still larger than the summed direct and pre-equilibrium contributions. The compound process dominates at the low neutron emission

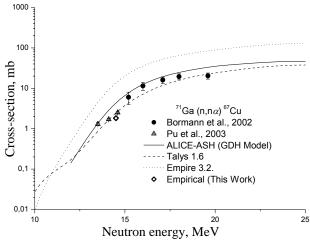


Fig. 7. The comparison of calculated excitation function of  $^{71}$ Ga(n,  $n\alpha$ ) $^{67}$ Cu reaction with the values reported in Ref. [42].

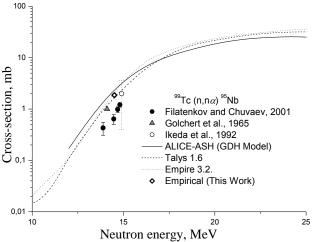


Fig. 9. The comparison of calculated excitation function of  $^{99}\text{Tc}(n, n\alpha)^{95}\text{Nb}$  reaction with the values reported in Ref. [42].

energy. Multiple compound emissions have been considered in this energy range. Talys pre-equilibrium calculations have been carried out with two-component exciton model.

Empire 3.2 calculation results are nearly similar with the ALICE/ASH and Talys 1.6 calculations at the compound reaction process. Theoretical calculations are tenfold above the other code results over 15 MeV incoming neutron energies in Figs. 6 and 7. Generally, results are close to empirical equivalents at the 14 - 15 MeV. It should be noted that all nuclear code results can be made better with the more suitable theoretical models and their parameters such as; mean free path, level density, deformation parameters, optical model parameters etc. Default and standard values have been selected for compound and pre-compound modelled calculations in this study.

Empirical calculations have been performed using the empirical formula in Table 1. These values have the best compatibility according to the experimental data which is taken from EXFOR and they are very close to theoretical code calculations.

#### 5. Conclusion

This paper has explored the dependence of the asymmetric properties of target nuclei with the cross-sections for  $(n, n\alpha)$  reactions. We have determined a different parameter groups by the classification of nuclei into even-even and odd-even. The excitation functions of a number of target nuclei of the  $(n, n\alpha)$  reactions have also been calculated utilizing pre-equilibrium and evaporation equilibrium nuclear models. The calculations were then compared with the experimental data. The results can be summarized and concluded as follows:

sult was shown in previous papers for different reactions. This consequence was confirmed with recent experimental data for the  $(n, n\alpha)$  nuclear reactions.

2. The reaction mechanism and particle emission

1. As the asymmetry parameter increases, cross-section values decrease for about 14 - 15 MeV. This re-

- 2. The reaction mechanism and particle emission depend strongly on the asymmetry parameter for the  $(n, n\alpha)$  cross-sections.
- 4. Odd (*Z*)-Even (*N*) systematics have a good fitting correlation ( $R^2$ ) value for (n,  $n\alpha$ ) cross-sections.
- 5. Parameter-based empirical relations are practical and applicable for  $(n, n\alpha)$  cross-section calculations at the 14 15 MeV neutron incident energies. Cross-section calculations are necessary for estimating unknown experimental data. The results of this and similar studies contribute to the analysis of cross-section characteristics. It is expected that the results also will help to determine the best empirical relation and its parameters by comparison with similar ones.

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## ОЦІНЕНА ЕМПІРИЧНА СИСТЕМАТИКА ПЕРЕРІЗІВ РЕАКЦІЇ $(n, n\alpha)$ ПРИ ЕНЕРГІЯХ 14 - 15 МеВ

У цьому дослідженні застосоване емпіричне рівняння, запропоноване Тел та ін., що включає параметр асиметрії для реакції  $(n, n\alpha)$  при енергіях 14 - 15 MeB. Значення вільних параметрів рівняння були визначені при підгонці експериментальних перерізів. Класифікація ядер на парно-парні та непарно-парні була прийнята для врахування ефекту спарювання на перерізи  $(n, n\alpha)$  реакції. Розрахунки функції збудження були проведені для таких ядер мішеней, як  $^{65}$ Cu,  $^{71}$ Ga,  $^{31}$ P і  $^{99}$ Tc, для  $(n, n\alpha)$  реакції при енергіях нейтронів, що налітають, до 30 MeB. Проведено порівняння між наявними експериментальними даними, розрахунками перерізів за ядерними моделями та результатами, обчисленими за емпіричними формулами. Визначені співвідношення для перерізів  $(n, n\alpha)$  реакції працюють при вхідних енергіях нейтронів 14 - 15 MeB. Зроблені розрахунки прийнятно узгоджуються з відомими експериментальними даними.

 $Ключові \ слова$ : емпіричні формули, параметр асиметрії,  $(n, n\alpha)$  реакції, систематика перерізів, програми для ядерних реакцій.

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## ОЦЕНЕННАЯ ЭМПИРИЧЕСКАЯ СИСТЕМАТИКА СЕЧЕНИЙ РЕАКЦИИ $(n, n\alpha)$ ПРИ ЭНЕРГИЯХ 14 - 15 МэВ

В этом исследовании применено эмпирическое уравнение, предложенное Тел и др., которое включает параметр асимметрии, для реакции  $(n, n\alpha)$  при энергиях 14 - 15 МэВ. Значения свободных параметров уравнения были определены при подгонке экспериментальных сечений. Классификация ядер на четно-четные и нечетно-четные была принята для учета эффекта спаривания на сечения  $(n, n\alpha)$  реакции. Расчеты функции возбуждения были проведены для таких ядер мишеней, как  $^{65}$ Cu,  $^{71}$ Ga,  $^{31}$ P и  $^{99}$ Tc, для  $(n, n\alpha)$  реакции при энергиях налетающих нейтронов до 30 МэВ. Проведено сравнение между имеющимися экспериментальными данными, расчетами сечений за ядерными моделями и результатами, вычисленными по эмпирическим формулам. Определенные соотношения для сечений  $(n, n\alpha)$  реакции работают при входных энергиях нейтронов 14 - 15 МэВ. Произведенные расчеты приемлемо согласуются с известными экспериментальными данными.

Ключевые слова: эмпирические формулы, параметр асимметрии,  $(n, n\alpha)$  реакции, систематика сечений, программы для ядерных реакций.

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