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Research Article

A Theoretical Study on the Production Cross–Section Calculations for ^{24}Na Medical Isotope

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Abstract

^{24}Na is a beta and gamma emitting radioisotope that is widely used in medical field as medical radiotracing. This radioisotope with a half-life of 14.977 hours is mostly produced in cyclotrons. The cross-section of a radioisotope is used to obtain various data about the production of that radioisotope. In cases where it is not possible to obtain experimental data, missing data can be completed with cross-section calculations and the obtained data can be compared with experimental, saving time, cost and effort. This study was carried out for detailed analysis of cross-section calculations for ^{24}Na isotope, which has a wide range of usage in medicine. In this direction, the cross-sections obtained from different computation programs were compared with literature data of the reactions. Production cross-sections of the ^{24}Na isotope were investigated in the ^{23}Na(d,p)^{24}Na, ^{24}Mg(n,p)^{24}Na, ^{25}Mg(n,n+p)^{24}Na, ^{25}Mg(p,2p)^{24}Na, ^{26}Mg(d,α)^{24}Na and ^{27}Al(n,α)^{24}Na reactions with the equilibrium and pre-equilibrium models of nuclear reaction codes of TALYS 1.95 and EMPIRE 3.2. Results were compared with experimental data from the EXFOR data library. The relevance of the models to the reactions was discussed and calculations were made using the relative variance analysis method to determine the most consistent model.

Keywords: Radioisotope, Cross-Section, TALYS 1.95, EMPIRE 3.2, Na-24

^{24}Na Tıbbi İzotopu için Üretim Kesiti Hesaplamaları Üzerine Teorik Bir Çalışma

Öz

^{24}Na, tıp ve mühendislik alanlarında yaygın olarak kullanılan beta ve gama yayan bir radyoizotoptur. Genellikle tıbbi radyo izlemede kullanılır. Yarı ömrü 15 saat olan bu radyoizotop, çoğunlukla siklotronlarda üretilir. Bir radyoizotopun tesir kesiti, o radyoizotopun üretimi hakkında çeşitli veriler elde etmek için kullanılır. Deneysel veri elde etmenin mümkün olmadığı durumlarda, eksik veriler tesir kesiti hesaplamaları ile tamamlanabilmekte ve deneysel verilerle karşılaştırılarak zamandan, maliyetten ve emekten tasarruf edilebilmektedir. Bu çalışma, tıpta ve endüstride geniş kullanım alanına sahip olan ^{24}Na izotopunun tesir kesiti hesaplamalarının detaylı analizi için yapılmıştır. Bu doğrultuda, farklı simülasyon programlarından elde edilen tesir kesitleri reaksiyonların literatür verileri ile karşılaştırılmıştır. ^{24}Na izotopunun üretim kesiti; ^{23}Na(d,p)^{24}Na, ^{24}Mg(n,p)^{24}Na, ^{25}Mg(n,n+p)^{24}Na, ^{25}Mg(p,2p)^{24}Na, ^{26}Mg(d,α)^{24}Na ve ^{27}Al(n,α)^{24}Na reaksiyonlarında TALYS 1.95 ve EMPIRE 3.2 nükleer reaksiyon kodlarının denge ve denge-öncesi modelleri ile incelenmiştir. Sonuçlar EXFOR veri kütüphanesinden alınan deneysel değerlerle karşılaştırılmıştır. Modellerin reaksiyonlara uygunluğu tartışılmış ve en iyi modeli belirlemek için göreli varyans analizi yöntemi kullanılarak hesaplamalar yapılmıştır.

Anahtar Kelimeler: Radyoizotop, Tesir Kesiti, TALYS 1.95, EMPIRE 3.2, Na-24

1. Introduction

Tracers are substances that enable tracking of various processes and systems. Radioactive tracers, also known as radiotracers, which can produced mainly in research reactors and accelerators, has an increasing importance in medical applications. They are used in a large proportion of diagnostic medicine, such as Single Photon Emission Computed Tomography

(SPECT), Positon Emission Tomography (PET) and Computed Radioactive Particle Tracking (CARPT) applications (IAEA, 2021).

The number of radionuclide types suitable for use as radiotracers has increased since the discovery of artificially produced radioactive isotopes. It was possible to produce on a wide scale of radioactive isotopes with the cyclotron and reactor. Radionuclides produced by the cyclotrons and reactors were used for biomedical research, some clinical trials and basic research in biochemistry. It became possible to produce radioactive isotopes of a number of biological elements. The first use of artificial radioactive sodium for medical purposes was in 1937. One of the most important uses of radioactive Sodium-24 (^{24}Na) is its use as a radiotracer in medicine and industry (IAEA, 2003; IAEA, 2008).

The use of various artificial radioactive elements as radiotracer has been reported in a significant number of biological and physiological studies. Unless the dose of the radioactive isotope is given large enough to affect living tissue, the result is the same as that obtained from the application of the stable isotope. Radioactive isotopes are used in many ways for peripheral vascular occlusion studies. The isotope to be applied is administered intravenously and the dose concentration in the periphery is measured. The method provides a simple way of determining blood flow to an extremity via major arteries or collateral circulation. Since the vitality of an extremity depends on its blood supply, it is clear that any information on this subject should be useful in evaluating the diagnosis, prognosis and treatment. In practice, observations obtained with this method have been helped to demonstrate the adequacy of collateral circulation, especially when the main arteries are partially or completely occluded (Quimby and Smith, 1944).

Each radioactive isotope emits beta or gamma, or both, during the decay process, and this can be detected by various ways. Some radioisotopes emit so penetrating radiation that their presence in various parts of the living organism can be detected and measured. One of these isotopes is radioactive ^{24}Na (γ_1=1.369 MeV, 99.9936 %; γ_2=2.7554 MeV, 99.855 %; β_1^-=1.393 MeV, 99.939 %; β_2^-=4.123 MeV, 0.003 %) (Smith and Quimby, 1947; IAEA, 2001). Radioactive ^{24}Na was first used in the diagnosis of peripheral vascular occlusions at Presbyterian Hospital in 1943. The first intramuscular clinical studies of ^{24}Na were conducted by Kety (1948). In some studies, the choice of this tracer is because it can spread easily in the tissue and has a short half-life ($t_{1/2}$=14.977 hours). The rate of cleaning depends on the tissue circulation and can be measured. This isotope has provided information with a high degree of accuracy in the fields of diagnosis, therapy and prognostic. This provides great benefits to the patient and the clinician (Smith and Quimby, 1947; Semple et al., 1951). Also noradrenalin sensitivity in hypertension has been measured with the ^{24}Na technique (Moulton et al., 1957)

Usage areas of radioisotopes have become widespread and analysis has gained importance thanks to the developing technology and scientific advances. One of the areas where radioisotopes are most commonly used is the medical applications. Many studies investigating the effects of different parameters of radioisotopes and their production routes, which have a wide range of uses in medical applications, have contributed to literature. (Özdogan, 2019; Şekerci, 2019; Şekerci, 2020a; Şekerci et al, 2020a; Şekerci et al, 2020b; Özdoğan et al, 2021a; Özdoğan et al, 2021b)

In this context, it is quite common to use different theoretical models to obtain various parameters that can inform researchers in cases where experimental studies cannot be carried out (Özdoğan et al, 2019a; Şekerci et al, 2019; Özdoğan, 2021; Özdoğan et al, 2021c; Özdoğan et al, 2021d). The cross-section value, which can be briefly explained as the probability of a reaction's occurrence, is also an important parameter for researchers. Considering all these, in this study, it is aimed to investigate the effects of some theoretical models in some of the production route reactions of ^{24}Na radioisotope, the importance of which is indicated in medical field and some models in production cross-section calculations. In the calculations, the TALYS code version 1.95 (Koning et al, 2019) and the EMPIRE code version 3.2 (Herman et al.,2007) were used and the results of the calculations were compared with the data from The Experimental Nuclear Reaction Data (EXFOR) library (Zerkin ve Pritychenko, 2018). The models, which are most compatible with the experimental data, were selected by the relative variance analyses (Kurenkov et al., 1999) method. There are many studies that contributed to the literature by examining the cross-section calculations of radioisotopes via various theoretical models and calculation tools (Kaplan et al, 2020; Özdoğan et al, 2020a; Özdoğan et al, 2020b; Şekerci et al, 2020c; Şekerci, 2020b; Karaman et al, 2020; Sarpün et al, 2019; Özdoğan et al, 2019b; Özdoğan et al, 2019c).

2. Calculation Methods

The cross-section means probability that two particles colliding. It is not possible to take measurements of some short half-lived nuclei. In such cases where the experimental data is insufficient or difficult to obtain, incomplete data can be completed by using nuclear reaction codes. It is also advantageous in terms of time, effort and cost (Şekerci and Kaplan, 2018). There are many calculation programs available to obtain the cross-section data of various reactions, where TALYS and EMPIRE are two of them, which are also used in this study.

TALYS 1.95 open source nuclear reaction analysis code was used to access nuclear reaction data which is developed by Netherlands, France, Belgium and Vienna cooperation. TALYS version 1.95 has been introduced in 2019. For targets which are in the 1 keV - 1 GeV energy range; it is used for the analysis and estimation of neutron, proton, deuteron, triton, ^{3}He and alpha induced nuclear reactions (Koning et al, 2019).

EMPIRE 3.2 is another cross-section calculation program which is also used in this study. EMPIRE is a computer program consisting of various nuclear models, nuclear reaction calculation codes, designed for calculations of energies and particles over a wide energy range. Deuterons, photons, nucleons, helions (^{3}He), tritons, alpha’s and light or heavy ions can be selected as projectiles. The possible energy range extends up to few hundred MeV for the induced partciles (Herman et al., 2007).

A nuclear reaction mechanism depends on the energy of the incoming particle. Compound nuclear processes are dominant in reactions with particles which have incident energy below 10 MeV. Hauser-Feshbach theory is widely used in the study of nuclear reactions that result in the decay of the composite nucleus into discrete and continuous states. In this study, equilibrium calculations have been acquired by using Hauser-Feshbach model.

In this study, the pre-equilibrium calculations have been obtained by using Two-Component Exciton Model. Pre-equilibrium models are very successful in explaining the high energy region of the energy spectrum in reactions with 10-60 MeV energies of protons, neutrons and alpha particles. However, these models are not successful in predicting the angular distributions of emitted particles. Pre-equilibrium mechanism, depending on the mass of the target nuclei and the exciton energy of the compound system plays a more important role in the emission of primary neutrons, protons and alpha particles compared to other reaction types (Millazzo-Colli et al., 1974).

Relative variance analysis used for choosing the most coherent model. The model with the smallest value found as a result of the calculations made with the formula given below as Equation 1, which is based on the comparison of the experimental data and the data obtained from the cross-section calculations, shows us the best model (Kurenkov et al., 1999).

Relative variance analysis method;

$$D = \frac{1}{N}\sum_{i=1}^{N}\left|S_i^{cal} - S_i^{exp}\right|/S_i^{exp} \quad (1)$$

3. Results and Discussion

In this study, in order to contribute to the development of the ^{24}Na radioisotope production routes, the production cross-sections of ^{23}Na(d,p)^{24}Na, ^{24}Mg(n,p)^{24}Na, ^{25}Mg(n,n+p)^{24}Na, ^{25}Mg(p,2p)^{24}Na, ^{26}Mg(d,α)^{24}Na and ^{27}Al(n,α)^{24}Na reactions have been examined up to 55 MeV via equilibrium and pre-equilibrium models with TALYS 1.95 and EMPIRE 3.2 nuclear reaction codes in where the graphical representations of the outcomes are given in Figs. 1-6. Calculated results were competed with experimental data taken from the EXFOR data library and the most relevant reaction and model for the production of ^{24}Na were determined by the relative variance analysis method. Relative variance analysis results have been given in Table 1. In addition, optimum production energy intervals and Q-values which represents change in energy of the reaction for the investigated reactions have been shown in Table 2 and Table 3 respectively.

In Fig. 1, ^{23}Na(d,p)^{24}Na reaction cross–section calculations against the data of Lange et al. (1969), Hirsh et al. (2015), Bowen et al. (1962) and Clarke et al. (1944) have been shown. While all models gave similar values up to 12 MeV deuteron energy, after that experimental data has higher values than model results as shown in Fig. 1. EMPIRE Exciton model is the most proper model for ^{24}Na production with 0.46478247 relative variance analysis result taken from Table 1. The optimum production energy interval for ^{23}Na(d,p)^{24}Na reaction is between 2 - 8 MeV as seen in Table 2.

Fig. 2, shows the cross-section calculations of ^{24}Mg(n,p)^{24}Na reaction. Experimental data taken from EXFOR, which are belongs to Mannhart et al. (2007), Geraldo et al. (1989), Ji-Zhou et al. (2089), Paulsen et al. (1965) and Butler et al. (1963). For 9–13 MeV energy region, Two-Component Exciton calculations have higher cross–section data than the experimental results. According to the analysis given in Table 1, 0.25766745 relative variance results show us EMPIRE Exciton is the best compatible model. As shown in Table 2, optimum production energy for ^{24}Mg(n,p)^{24}Na reaction is between 10 – 15 MeV.

In Fig. 3, ^{25}Mg(n,n+p)^{24}Na reaction cross–section calculations shown. Experimental data belongs to Ikeda et al. (1988). TALYS Hauser-Feshbach is the most correlative model with 0.68906043 relative variance result. Looking at the Table 2, the optimum production energy for ^{25}Mg(n,n+p)^{24}Na reaction can be seen as 13.5 – 15 MeV interval.

^{25}Mg(p,2p)^{24}Na reaction cross–section calculations are shown in Fig. 4. The experimental data are taken from Cohen et al. (1954). Although the TALYS Two-Component Exciton model up to 19 MeV follows the experimental data from below, it is in good agreement with the experimental data after this energy. According to the analysis given in Table 1, TALYS Two-Component Exciton is the most consistent model with 0.21808735 relative variance analysis result. For ^{25}Mg(p,2p)^{24}Na reaction, optimum production energy is in the range od 20 – 22 MeV, which can also be seen from Table 2.

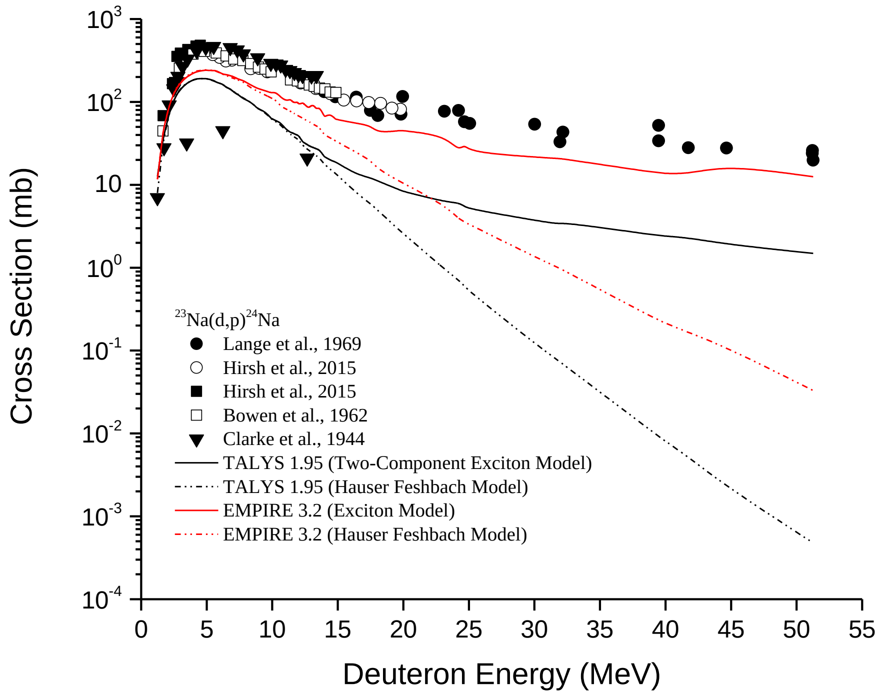

Fig. 1. Cross-sections calculations of the $^{23}Na(d,p)^{24}Na$ reaction

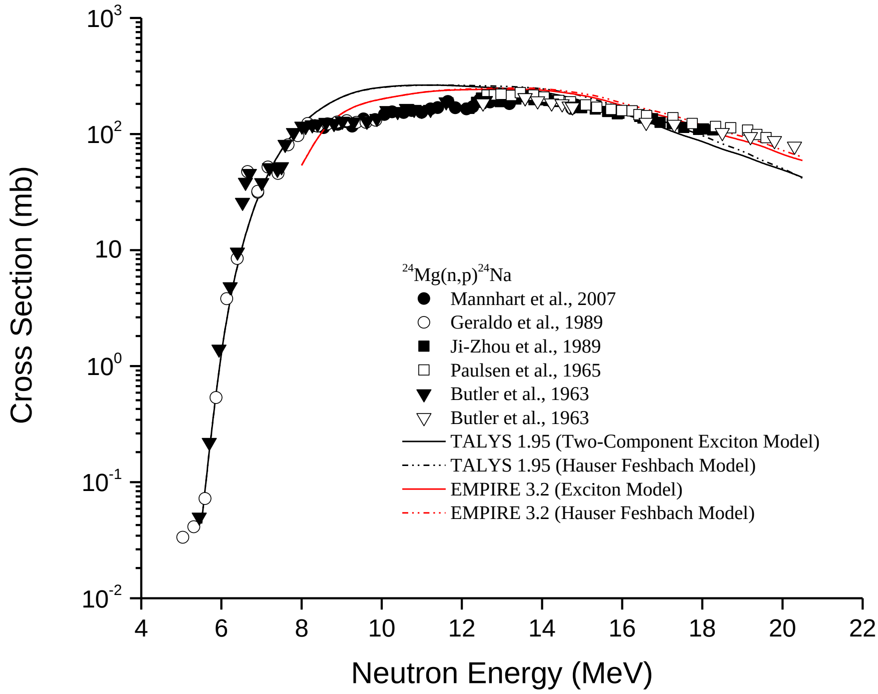

Fig. 2. Cross-sections calculations of the $^{24}Mg(n,p)^{24}Na$ reaction

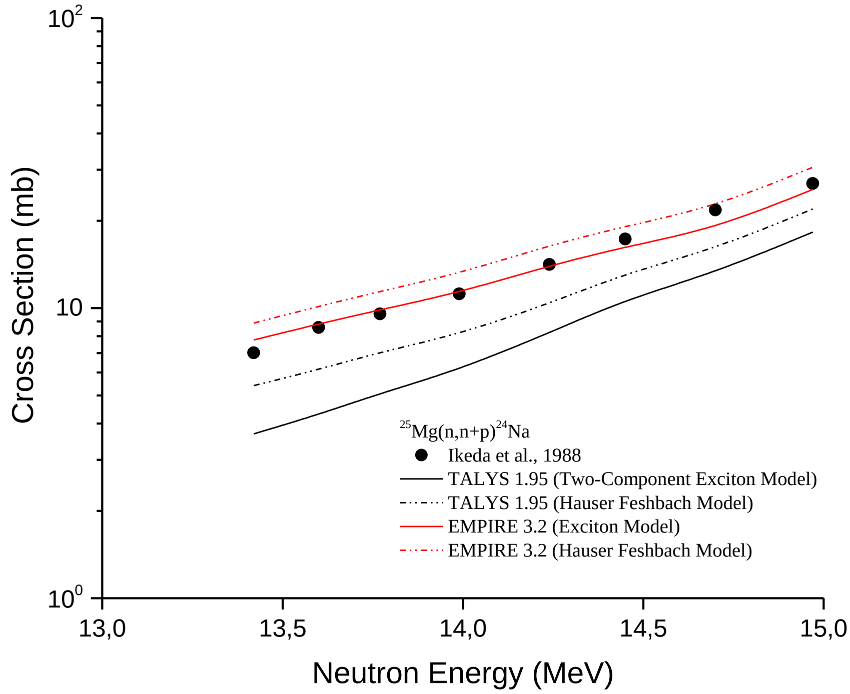

Fig. 3. Cross-sections calculations of the $^{25}Mg(n,n+p)^{24}Na$ reaction

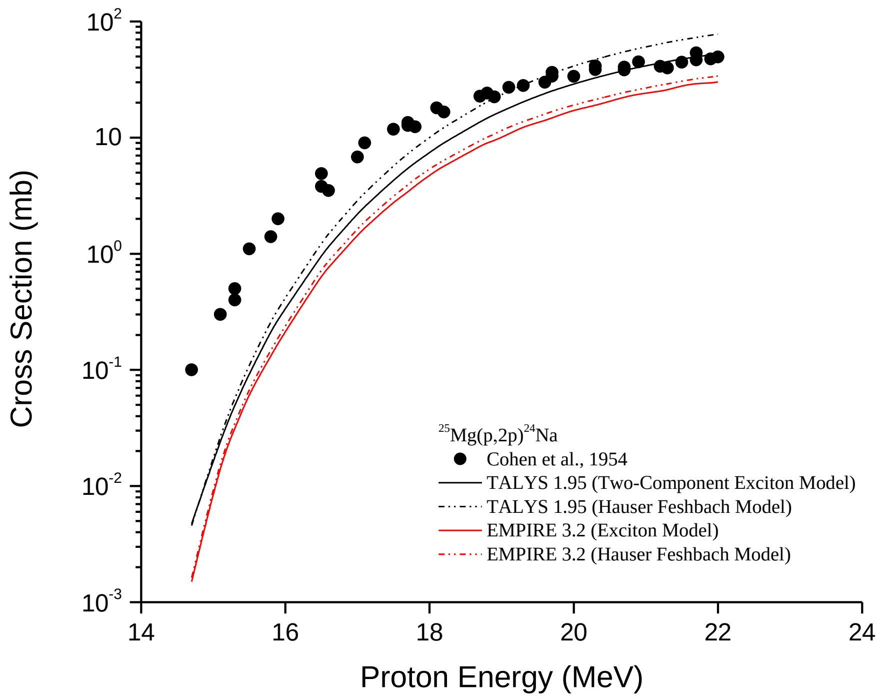

Fig. 4. Cross-sections calculations of the $^{25}Mg(p,2p)^{24}Na$ reaction

$^{26}Mg(d,\alpha)^{24}Na$ reaction production cross–section calculations have been given in Fig. 5, which the experimental data taken from Roehm et al. (1969). While all calculations in good agreement with the experimental data up to 11 MeV, after this energy none of the models are seems to be compatible with the experimental data. According to the relative variance analysis results, EMPIRE Exciton is the most coherent model for this reaction. From Table 2, the optimum production energy for the ^{26}Mg(d,α)^{24}Na reaction can be seen as 8 – 12 MeV interval.

In Fig. 6, ^{27}Al(n,α)^{24}Na reaction cross–section calculations have been given against the experimental data which have been obtained from Uno et al. (2007), Menlove et al. (2007), Grundl et al. (2007) and Mannhart et al. (2007). Two-Component Exciton calculations are more compatible with the experimental results for 6 – 15 MeV energy region, while Exciton model is more harmonious for 15 – 30 MeV energy region. According to the Table 1, TALYS Two-Component Exciton is the best model for ^{27}Al(n,α)^{24}Na reaction with 0.09355778 relative variance results. 8 – 12 MeV is the optimum production energy interval for this reaction.

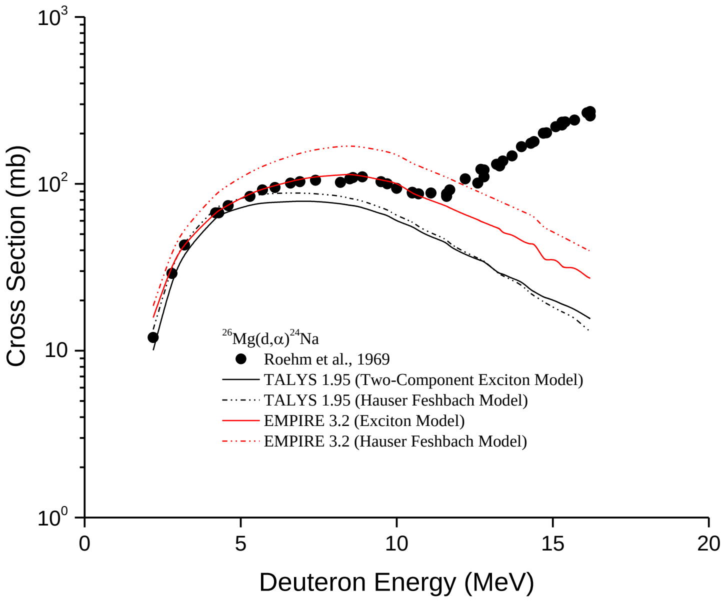

Fig. 5. Cross-sections calculations of the ^{26}Mg(d,α)^{24}Na reaction

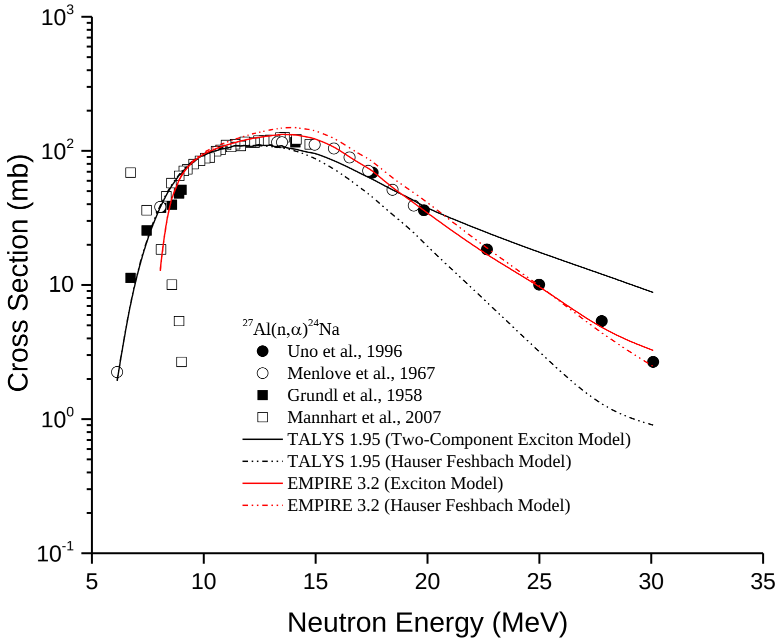

Fig. 6. Cross-sections calculations of the ^{27}Al(n,α)^{24}Na reaction

Table 1. Relative variance analysis of ^{24}Na production cross–section calculations

Reactions	TALYS Two-Component Exciton	TALYS Hauser-Feshbach	EMPIRE Exciton	EMPIRE Hauser-Feshbach
$^{23}Na(d,p)\,^{24}Na$	0,678833	0,688679	0,464782	0,546517
$^{24}Mg(n,p)\,^{24}Na$	0,315686	0,332176	0,257667	0,274235
$^{25}Mg(n,n+p)^{24}Na$	0,695673	0,68906	0,693677	0,762526
$^{25}Mg(p,2p)^{24}Na$	0,218087	0,370652	0,504497	0,445446
$^{26}Mg(d,\alpha)\,^{24}Na$	0,63995	0,622437	0,487062	0,562844
$^{27}Al(n,\alpha)\,^{24}Na$	0,093558	0,133723	0,100691	0,191011

Table 2. Optimum energy of ^{24}Na production

Radioisotope	Production Reaction	Optimum Energy Interval (MeV)
^{24}Na	$^{23}Na(d,p)\,^{24}Na$	2 – 8
^{24}Na	$^{24}Mg(n,p)\,^{24}Na$	10 – 15
^{24}Na	$^{25}Mg(n,n+p)^{24}Na$	13.5 – 15
^{24}Na	$^{25}Mg(p,2p)^{24}Na$	20 – 22
^{24}Na	$^{26}Mg(d,\alpha)\,^{24}Na$	8 – 12
^{24}Na	$^{27}Al(n,\alpha)\,^{24}Na$	11 – 18

Table 3. Q-values of ^{24}Na production

Radioisotope	Production Reaction	Q-values (MeV)
^{24}Na	$^{23}Na(d,p)\,^{24}Na$	4.73480
^{24}Na	$^{24}Mg(n,p)\,^{24}Na$	4.73331
^{24}Na	$^{25}Mg(n,n+p)^{24}Na$	-9.83928
^{24}Na	$^{25}Mg(p,2p)^{24}Na$	-12.0639
^{24}Na	$^{26}Mg(d,\alpha)\,^{24}Na$	2.91417
^{24}Na	$^{27}Al(n,\alpha)\,^{24}Na$	-3.13255

4. Conclusions

In this study; production cross-sections of ^{24}Na radioisotope, which is used in medical applications, was investigated with the help of equilibrium and pre-equilibrium nuclear reaction models. Equilibrium and pre-equilibrium states of the isotope were modeled with TALYS 1.95 and EMPIRE 3.2 nuclear reaction codes and compared with the experimental data. Two-Component Exciton Model was used in the TALYS 1.95 program for the pre-equilibrium state, while the Hauser-Feshbach model was used for the equilibrium state. Similarly, in EMPIRE 3.2 program, Exciton Model was used for pre-equilibrium state, while Hauser-Feshbach model was used for equilibrium state. Experimental data were obtained from EXFOR data library of the International Atomic Energy Agency. The most suitable model for each reaction was chosen with the relative variance analysis calculations.

It can be said by considering the investigated reactions in this study that the cross-section calculation results obtained by pre-equilibrium models are in better harmony with the experimental data rather than the equilibrium models. Hauser-Feshbach models, which are equilibrium models, were not compatible with the experimental results.

EMPIRE Exciton model is the most consistent model for the ^{23}Na(d,p) ^{24}Na, ^{24}Mg(n,p)^{24}Na and ^{26}Mg(d,α) ^{24}Na reactions. TALYS Two-Component Exciton model is the most coherent model for ^{25}Mg(p,2p)^{24}Na and ^{27}Al(n,α) ^{24}Na reactions, while TALYS Hauser-Feshbach is the most proper model for ^{25}Mg(n,n+p)^{24}Na reaction. Model parameters of the reactions examined in this study can be revised for more accurate results with the experimental data.

When the optimum production energies of the reactions are given in Table 2, almost 5 MeV deuteron energy is enough for the production of ^{24}Na radioisotope via ^{23}Na(d,p) ^{24}Na reaction.

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