INVESTIGATION OF THE REACTION CROSS-SECTION FOR PRODUCTION THE RADIOACTIVE ISOTOPES USED IN FABRICATING THE NUCLEAR BATTERIES

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Abstract. Beta-decay radioactive isotopes have been widely used as a high-energy source in nuclear batteries. The aim of this study is to improve the evaluations of nuclear reactions for the production of some radionuclides with possible use in nuclear batteries. In particular, the 38Ar(n,γ)39Ar, 59Co(n,γ)60Co, 112Cd(n,γ)113mCd, 151Eu(n,γ)152Eu, 153Eu(n,γ) 154Eu, 193Os(n,γ) 194Os and 203Tl(n,γ) 204Tl neutron capture reactions have been studied for an incident neutron energy range from 10^{-6} eV up to 2 MeV. The evaluations of the capture cross sections are based on the nuclear model calculations with the latest release code EMPIRE-3.2.2 (Malta) and compared with the available experimental ones. The TENDL nuclear data library, which provides the output of TALYS nuclear model code system have been also presented along with the extracted data for some interactions with no experimental support. Simulation of the beta particles transportation and the generated bremsstrahlung intensity through silicon carbide semiconductor has been also investigated.

Keywords: Nuclear batteries; Nuclear cross section; Neutron capture reactions.

1. Introduction

Due to the remote environmental conditions and the growth of long-range space missions and sea voyages and many other different fields such as human-portable energy sources, robotic sensors, developing fields and military missions, the need for alternative energy sources with special specifications, especially longevity ones, has increased. To serve this role Moseley and Harling in 1913 [1] have been used the natural radioactive isotope as a substitutional source of energy to the chemical once in what is called the nuclear batteries. These batteries with a good shielding and design can uniquely save much more energy than their chemical counterparts, which can be close to 1,000 watts per hour and the life of it can be in the order of many decades since it is related to the half-life of the radioisotope used. With the availability of low-energy electronics and the fact that radioactive sources are inexpensive, nuclear batteries can be considered as a source of commercial profit as well as a source of high energy [2].

There are many competing types of nuclear batteries: thermoelectric, thermophotoelectric, direct charge collection, thermionic, scintillation intermediate, alphavoltaics, and betavoltaics. Between these types, the most interested ones for the past four decades is the radioisotope thermoelectric generator (RTG) in which the heat produced by the isotopes decay converted to electricity through the Seebeck effect [3, 4]. Selection of radioisotopes must fulfill a good match between the range of radiation and the scale length of the transducer, which in turn depends on the decay type of the radioisotopes. It also depends on the cost of the production and separation of the radioisotopes. The alpha and beta emitters are the most types of radioisotopes used in nuclear batteries. In the present work, the emphasis was
placed on the nuclear reactions that produce the isotopes with beta particles decay, which have a longer range than the alpha emitters have and can be used in different types of batteries.

The nuclear reaction mechanisms for the nuclear isotopes used in the nuclear batteries can be understood from the information provided by the calculation of the nuclear reaction cross-sections, which is also needed in many other applications such as fusion reactor and nuclear power plants. This study is an attempt to evaluate the cross-sections for some nuclear reactions, which produce radionuclides used in nuclear batteries with the code EMPIRE [5]. Many nuclear models for cross-sections calculations have been included in this complex and flexible system and designed for calculations over a broad range of energies and incident particles.

The reaction of an incident particle with an atomic nucleus can take place in many ways. In studies of light-ion induced nuclear reactions, one distinguishes between three different mechanisms: direct, compound and pre-equilibrium nuclear reactions, which can be subdivided according to time scales or, equivalently, the number of intra nuclear collisions taking place before emission. Furthermore; each mechanism preferentially excites retain parts of the nuclear level spectrum and is characterized by different types of angular distributions.

2. Nuclear reaction cross-section

The nuclear reaction mechanisms of an incident particle with an atomic nucleus can be explained by the three main models or mechanisms mentioned above according to the energy of the projectile and time scales of the interaction. At lower excitation energies (below 10 MeV), the whole system is involved in the interaction and the isotropic distributions of emitted particles in the center of the mass frame can be explained by the compound nucleus mechanism. In this model the theoretical calculations of reaction cross-sections are described by Hauser and Feshbach [6] taken into account the conservation of angular momentum and parity.

\[
\sigma_{ab}^{HF}(E) = \pi \lambda_a^2 \sum_{J; \pi} \frac{2J+1}{(2J+1)(2\pi)} T_a T_b \pi e^{-\frac{\lambda_a}{2}}
\]

(1)

where \(J; \pi\) are the total angular momentum and parity of the compound state, \(\lambda_a\) is the reduced wavelength in the incident channel (inverse of the wave number \(k\)) and \(T_a T_b\) are the transmission coefficients of the incident and outgoing channels respectively.

On the other hand, at relatively higher excitation energies, the forward peaked angular distribution of particles indicates the presence of direct reaction mechanism [7, 8] in which only a few nucleons take part in the evolution of the reaction processes. The direct interactions analysis procedure either permanently with the Distorted Wave Born Approximation (DWBA) or by the coupled channels approaches [9].

The theory of nuclear reactions was extended to include a statistical treatment for particles emitted from the target nucleus during the equilibrium in a process called pre-equilibrium nuclear reaction. Quantum mechanical and phenomenological theories were proposed for pre-equilibrium emission mechanisms such as the classical exciton model including nucleon, cluster and gamma emissions, and Blann’s Hybrid model that is limited to nucleon emission for the semiclassical theories. For quantum mechanics, the Feshbach, Kerman, and Koonin (FKK) model [10, 11] and Nishioka, Verbaarshot, Weidenmuller and Yoshida (NVWY) model are the most important models. For the evolution of the projectile-target system from small to large energy losses it is useful to consider separately the states with at least one particle in the continuum which is described in the Multi Step Direct (MSD) theory and the states with all particles are bound, i.e. the Multi Step Compound (MSC) reactions [12, 13].

The multi-step direct cross-sections were defined as the summation of n-step as follows:
\[
\frac{d^2\sigma}{d\Omega dE} = \sum_n \frac{d^2\sigma^{(n)}}{d\Omega dE}
\]

where the contribution of the n-step cross-sections can be given as a function of probability per energy to find the system in the configuration \(c, c=npnh\), and the open channel T-matrix:

\[
\frac{d^2\sigma}{d\Omega dE} = \sum_{c=[npnh]} P_c(E) \left| T_{c0}^{(n)} \right|^2
\]

with

\[
P_c(E) = -\frac{1}{\pi} \text{Im} \left[ \int dE' \ g(E-E')(c|G^{\text{in}tr}(E')|c) \right]
\]

The Lippman-Schwinger formalism gives for the nth order (= nth step) transition amplitude

\[
T_{\gamma\gamma'}^{(n)} = \langle X_E^{(c-)} | (y|V^{\text{res}}(G^{\text{chan}}(E)V^{\text{res}})^{n-1}|0|X_0^{(c+)})
\]

where \(X_E^{(-)}\), \(X_E^{(+)}\) and \(G^{\text{chan}}(E)\) are the incoming and outgoing scattering waves and the Green’s function for the channel \(y\) respectively.

\(V^{\text{res}}\) represent the residual effective projectile-target interaction which gives the effective Hamiltonian with the averaged energy Hamiltonian of the optical model \(H^{\text{opt}}\) and intrinsic Hamiltonian \(H_{\text{intr}}\) of the asymptotically separated nuclei.

Since the multi-step theory is considered in the EMPIRE code for two-step reactions only, then \(n\) must equal 2, and the cross-section distribution for the second step is:

\[
\frac{d^2\sigma^{(2)}}{d\Omega dE} = \sum_{\lambda_1\lambda_2} \int dE_1 S_{\lambda_2}(E,E_1) S_{\lambda_1}(E_1,0) \left| \frac{d\sigma^{(2)}}{d\Omega}(E,E_1) \right|_{\lambda_1\lambda_2}
\]

\(S_{\lambda_2}\), the transition strength function and \(\lambda\) is the momentum transfer.

According to NVWY, the average MSC cross-section leading from the incident channel \(a\) to the exit channel \(b\) is given by:

\[
\frac{d\sigma_{ab}}{dE} = (1 + \delta_{ab}) \sum_{n,m} T_n^a \Pi_{n,m} \Pi_m^b \]

where \(\Pi_{n,m}\) is the probability transport matrix.

### 3. Reaction Modeling

The neutron induced reactions cross-sections on the target nuclei; \(^{38}\)Ar, \(^{59}\)Co, \(^{121}\)Cd, \(^{151}\)Eu, \(^{153}\)Eu, \(^{193}\)Os, and \(^{203}\)Tl targets have been calculated with the latest modular system EMPIRE 3.2.2 Malta [14]. The direct cross-section has been performed with optical model calculations using the ECIS06 code incorporated into the EMPIRE system. The option (DIRECT=1), considering a spherical optical model, were used to calculate the particle transmission coefficients for the emerging channels, while the formalism of the coupled-channel (CC) have been used for transmission coefficients calculations in the incident neutron channel. For non-coupled levels, the direct neutron scattering was considered by a DWBA with dynamic deformations selected to describe available neutron emission spectra. Quantum-mechanical preequilibrium models based on multi-step direct and multi-step compound were taken into account by (MSD and MSC) modules to consider the preequilibrium emission, which are suitable for calculation below 30 MeV of neutron incident energy. The compound nucleus mechanism was described by Hauser-Feshbach statistical model [6].
The global Koning and Delaroche [15] optical potential parameters taken from the Recommended Input Parameter Library (RIPL) have been tested for selected nuclear reactions. This is a dispersive and spherical OMP reproducing all available nucleon scattering data on target nucleus from 0.0 up to 200 MeV. The nuclear level densities can be calculated with the microscopic formalism (Hretree-Fock-Bogoliubov Model HFB) [16]. The parametrization of Gamma Ray Strength Functions (GSF) is particularly important for the study of capture reactions. In our calculations, we used the modified Lorenzian radiative-GSF (MLO) (Plujko MLO RIPL-2) [17] and width fluctuation correction was selected up to 3.00 MeV (for neutron induced) to account the correlation between the incident and exit channels in elastic scattering.

4. Results and Discussion

EMPIRE is a modular system designed for cross section calculations over a broad range of energies starts just above the resonance region of a neutron projectile, this appear clearly in the presence calculations below as a high deviation from the experimental data at very low energy (usually E<10^{-5} MeV). In addition, the cross section calculations of all the capture reactions investigated in this study are not reproducing very well in the resonance region, since the resonance module of the code requires that electronic version of the Atlas be installed. The copy rights for Atlas belong to the publisher (Elsevier), so we will not be able to install Atlas and therefore to use the resonance module of EMPIRE.

The theoretical calculated cross-section for the $^{38}\text{Ar}(n,\gamma)^{39}\text{Ar}$ reaction were illustrated in Fig. 1(a) along with the available experimental data [18, 19] obtained from the EXFOR library [20]. Inspection of these curves revel that the calculated results are overestimated the data from the TENDL library with the same behavior for all energy range with high deviation at very low energy. The deviation decreases gradually with increasing of the neutrons energies until it reaches approximately the limit of (1%). It can be also seen from this figure that the two points represented the experimental data are coincide with TENDL data. The cross-sections for $^{112}\text{Cd}(n,\gamma)^{113}\text{mCd}$ reaction are shown in Fig. 1(b). The calculated results for this reaction have been presented with the only two experimental points available from the EXFOR database.

Figure 1. Comparison of available experimental data and calculated neutron-capture cross-sections for the target nuclei (a) $^{38}\text{Ar}$ and (b) $^{112}\text{Cd}$.
In Fig. 2, $^{59}$Co(n,$\gamma$)$^{60}$Co nuclear reaction cross-sections have been calculated and compared with the available experimental data taken from Refs. [21-25] and with the TENDL data [26]. It is obvious that the calculated results are in good agreement with the experimental data in the neutron energy range 10^{-2}-10^1 MeV. At low energy range up to 10^{-2} MeV, the calculated data overestimate the experimental data. In this case, the best theoretical estimation is given by TENDL-2017 which coincides with the experimental data for energy range up to 10^{-2} MeV.

![Figure 2. Comparison of available experimental data and calculated neutron-capture cross-sections for the $^{59}$Co target nucleus.](image)

The nuclear reactions $^{151}$Eu(n,$\gamma$)$^{152}$Eu and $^{153}$Eu(n,$\gamma$)$^{154}$Eu are examined in Fig. 3. (a) and (b). The experimental cross-section values [27-32] for these interactions have shown a perfect agreement with the calculated results for all neutron energy region of 10^{-6}-10^0 MeV. The data from the TENDL library have been also shown in these figures for the two reactions mentioned above along with the experimental data and theoretical curves. Regarding to the $^{193}$Os(n,$\gamma$)$^{194}$Os reaction, the calculated results are depicted in Fig. 3 (c) along with the available experimental data [19, 33]. As can be seen from this figure, the calculated results are coinciding with the TENDL data in the neutron energy range 10^{-1}-10^1 MeV. Whereas at very high energy from 10 up to 100 MeV, the calculated results underestimate the TENDL data. In Fig. 3. (d) the cross-section of the $^{203}$Tl(n,$\gamma$)$^{204}$Tl reaction are presented together with the data from the TENDL library and compared with the experimental data [34-37]. Both results are in reasonable agreement.

In nuclear battery designs, the choice of a suitable radioisotope with a suitable energy of the ionizing radiation is very important. It is depending on the range of the energy emitted from the isotope in the material that the isotope is embedded in. for betavoltaic a p-n junction in a semiconductor is used as a transducer. Beta particles transfer energy to the target material electrons by Coulomb scattering and Bremsstrahlung emission [38]. Since the incident beta particles have a mass equal to the mass of the target electrons, their path of losses energy in a material will be random and undergoes significant scattering. Simulation of the beta particle tracks emitted from isotopes, used in this study, in SiC semiconductor have been investigated and illustrated in Fig. 4 using the PENYLOPE Monte Carlo code system [39]. In these figures we illustrate the YZ section were the beam is coming parallel to the z-axis vertically down on the sample. The energy of the beta particles
used in this simulation have been chosen with the maximum \( (E_{\text{max}}) \) energy of the emitted spectrum and listed in Table 1.

![Graphs](image)

**Figure 3.** Comparison of available experimental data and calculated neutron-capture cross sections for the target nuclei (a) \(^{151}\text{Au}\), (b) \(^{153}\text{Au}\), (c) \(^{193}\text{Os}\) and (d) \(^{203}\text{Tl}\).

**Table 1.** Characteristics of beta-emitting radioisotopes used in the study [38].

| Nuclide | Half-life (Years) | \( \beta_{\text{max}} \) (MeV) |
|---------|------------------|-------------------------|
| Ar-39   | 269              | 0.565                   |
| Co-60   | 5.2713           | 0.318                   |
| Cd-113m | 14.1             | 0.58                    |
| Eu-152  | 13.54            | 1.818                   |
| Eu-154  | 8.592            | 1.845                   |
Os-194  
6  
0.0966  

Tl-204  
3.78  
0.763
Due to the difference in the energies of the beta particles one can observe that the beta particles in SiC ranging from (maximum 3.0 mm for $^{154}$Eu source) to (minimum 0.04 mm for $^{194}$Os source). The backscattering beta particles have been also shown in these figures with a red path.

The generation of secondary gamma photons from bremsstrahlung events, produced from interaction of beta particles with nuclei, require an additional shielding in battery design \[40\]. Bremsstrahlung energy generation is effective when transducer material has a high atomic number since its scales approximately as $Z^2$ of the target \[41\]. In this paper, the silicon carbide semiconductor has been used as a transducer material with relatively low atomic number so that bremsstrahlung spectrum can be neglected. The probability density of the generated bremsstrahlung spectrum has been shown in Fig. 5 for beta particle sources under investigation using the PENELOPE code system \[42\]. It can be seen from the figures that the probability density is very small and decreases with increasing bremsstrahlung energy and it is approximately equal to zero at high energy photons. In these figures, the absorption energies, elastic scattering parameters and cutoff energies have been used as a default simulation values.
5. Conclusions

In the present work, the cross-sections for some reactions produced $\beta$ sources used in nuclear batteries have been calculated at spectrum averaged energies from $10^6$ eV up to 2 MeV. The obtained data for cross sections provide more responsible estimation values for these reactions and can be considered as additional data for nuclear data evaluation, which can be exercised in the different application fields of nuclear energy as well as in a testing of nuclear reaction models. It should be mentioned that the resonance model should be activated to representing cross sections of the neutron capture interactions in the low energy region. For nuclear batteries design the scale length for the materials contained the isotopes have to be in order of the mean free path of the radiation so that the optimum amount of energy contained in the radiation can be deposited in the transducer which is in this study is 0.04 mm is for $^{194}$Os and gets much larger as the beta energy increases.

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