Transition Metal Dichalcogenide Transmutation through Neutron Irradiation, Case Study: ZrS_2

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Abstract: The transmutation of the ZrS_2 Transition Metal Dichalcogenide (TMDC) layered materials through Neutron Irradiation is investigated and discussed. The study is performed by implementing the General Monte Carlo N-Particle (MCNP 6) computational tool. A model incorporating the standard linear attenuation coefficient experimental setup has been designed and validated with previously reported work. The results reveal that, under the neutron interaction rates, the Zr-92, Zr-94 and S-34 are neutron transparent. Additionally, the results resolve that the ZrS_2 TMDC can withstand the neutron radiative environment. Finally, the (n, alpha) interaction rate showed a higher threshold of about 2 MeV than most of the neutron energy spectrum in a thermal nuclear reactor.

Key words: Transition metal dichalcogenide (TMDC), radiation materials science, Monte Carlo N-Particle (MCNP), quasi two-dimensional materials.

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1. Introduction

The family of the transition metal dichalcogenides (TMDCs) represents an essential class of layered materials. They possess remarkable physical properties due to their quasi two dimensional (2D) layered type structures with massive potential in multiple applications such as high mobility field effect transistors, photovoltaic applications, energy storage, and optoelectronic applications [1]-[3]. Furthermore, environmentally friendly abundant, non-toxic and cheap materials are of high importance for industrial applications. TMDCs from groups IVb, Vb, and VIb display the whole spectrum of electronic properties covering the range from metals to large and narrow bandgap semiconductors and insulators. Their layered-type structures consist of a sheet of metal atoms sandwiched between two sheets of chalcogens. The bonding inside the layers is of strong mixed covalent-ionic character, while relatively weak Van der Waals (VdW) forces hold the adjacent sheets together [4], [5]. Zirconium disulfide (ZrS_2) belongs to the group IVb of TMDCs with transition metal Zr and chalcogens S.

Several research groups have reported the implementation of the ZrS2 for atomic layer deposition required in optoelectronic applications, e.g. [1], [4]. The ZrS_2 showed intense competition with MoS_2 in optoelectronic applications such as photodetectors. Optoelectronic applications in nuclear power industries have been widely studied since the 1990s [6]. In recent years, photodetectors are considered one of the most exciting applications of the optoelectronic technology used in monitoring applications in the nuclear power industry [6]. Additionally, Walker *et al*. [7] investigated the radiation effects on the two dimensional (2D) materials' structure and electronic performance. The study investigated the radiation tolerance of such materials in a radiative environment such as space and or nuclear reactors. In earlier work on radiation effects of TMDCs, the total ionizing dose effects on the TMDC, used in field effect transistors (FET), is reported by Zheng *et al*. [8]. The study presents them as future potential candidates for space applications.

Moreover, Vogl *et al*. [9] studied the tolerance of the 2D materials for space applications. The purpose of the current work is to study the transmutation of the ZrS_2 in a neutron radiative environment for photodetector monitoring applications in a neutron radiative nuclear reactor environment. The behaviour of ZrS_2 in an irradiative neutron environment is presented and discussed. This is performed by considering its transmutation due to neutron irradiation. The study should pave the way for a more comprehensive understanding of its material composition in an irradiative neutron environment and to what extent ZrS_2 can withstand the irradiative neutron environment. The General Monte Carlo N-Particle (MCNP 6) computational tool has been employed in order to calculate the neutron interaction rates for both zirconium (Zr) and sulfur (S).

Table 1. Chemical Composition of the Validation Sample

Material	PbO	B_2O_3	Na_2O	Li_2O	SiO_2
Mol %	5	40	25	5	25

2. Model Development and Simulations

In this work, the MCNP6 [10] computational tool is used to calculate the neutron interaction rates for Zr-92 and S-34 isotopes in the ZrS_2 TMDC materials. The code deals with the transport of neutrons, gamma rays, and coupled transport. It can be used in several transport modes: neutron only, photon only, electron, combined neutron/photon transport where the photons are produced by neutron interactions, neutron/photon/electron, photon/electron, or electron/photon. The neutron energy regime is from 10-11 MeV to 20 MeV, and the photon and electron energy regimes are from 1 keV to 1000 MeV. The standard experimental setup used in mass attenuation coefficient calculations has been modelled accordingly [10], [11]. Fig. 1 illustrates a schematic diagram of the employed model. It consists mainly of three parts, where the sample located between the point isotropic neutron source enclosed in a lead cylinder with a beam opening (on the left) and a sodium iodide detector enclosed in a lead cylinder with a beam opening corresponding to the sample. The (n. gamma) interaction rates based on the average flux in the sample cell obtained from MCNP 6 can be calculated using Equation (1) below.

$$\textit{Interaction Rate} = \emptyset * \Sigma * V_{ol}, \tag{1}$$

where ∅ is the average neutron flux over the ZrS_2 sample cell, Σ is the (n, gamma) macroscopic cross-section for ZrS_2, and Vol is the volume of the sample cell.

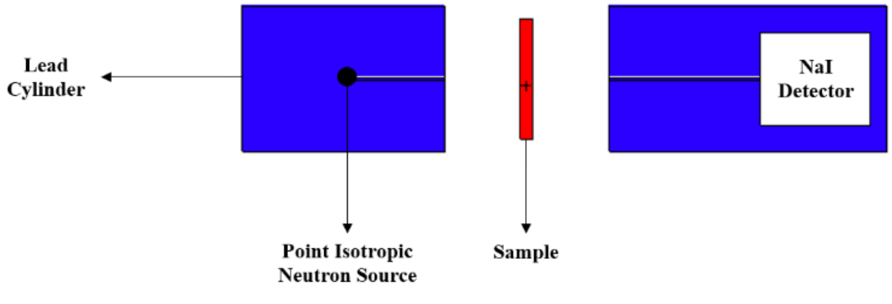

Fig. 1. MCNP6 model used for radiative capture interaction rates calculations. From left to right: Point isotropic neutron source enclosed in a lead cylinder with a beam opening, the sample, and a sodium iodide detector enclosed in a lead cylinder with a beam opening corresponding to the sample.

The performed simulation model has been validated throughout the calculations of the linear attenuation coefficient obtained for borosilicate glass samples according to work reported by Salama *et al.* [12]. The samples consist of the borosilicate glass, which includes lead, as listed in Table 1. The model's validation shows the relative error of less than 15% compared to the mass attenuation coefficient measured experimentally for Cs-162 0.662 MeV gamma line, as summarized in Table 2.

Table 2. MCNP6 Model Validation Metrics

Material	Theoretical	Experimental	MCNP6 Model	Relative Error %
Mass Attenuation Coefficient (cm^2/gm)	0.077	0.07±0.002	0.088±0.045	14.29%

3. Results and Discussion

Thorough knowledge of electromagnetic and particle irradiation effects on two-dimensional materials is crucial for an in-depth understanding of materials' electronic and physical properties for use in several applications. Ionizing radiation creates defects that could impact the structure and the electronic performance of the materials. The determination of these defects' impact is vital for developing 2D materials -based devices for use in high-radiation environments, such as space or nuclear reactors. One of the main reasons behind studying neutron radiative capture interaction rates with Zr-92 and Zr-94 is that their yield decay into Nb-93 and Mo-96. The decay can be seen according to Equations 2 and 3, respectively. Additionally, the S-34 yield (S-35) decays to Cl-35, which is clear to be observed according to Equation (4)

$$n + Zr - 92 \rightarrow Zr - 93 \xrightarrow{\beta -, T_{\frac{1}{2}} = 1.53*10^6\ years} Nb - 93, \tag{2}$$

$$n + Zr - 94 \rightarrow Zr - 95 \xrightarrow{\beta -, T_{\frac{1}{2}} = 64.05\ days} Mo - 95, \tag{3}$$

$$n + S - 34 \rightarrow S - 35 \xrightarrow{\beta -, T_{\frac{1}{2}} = 87.37\ days} Cl - 35, \tag{4}$$

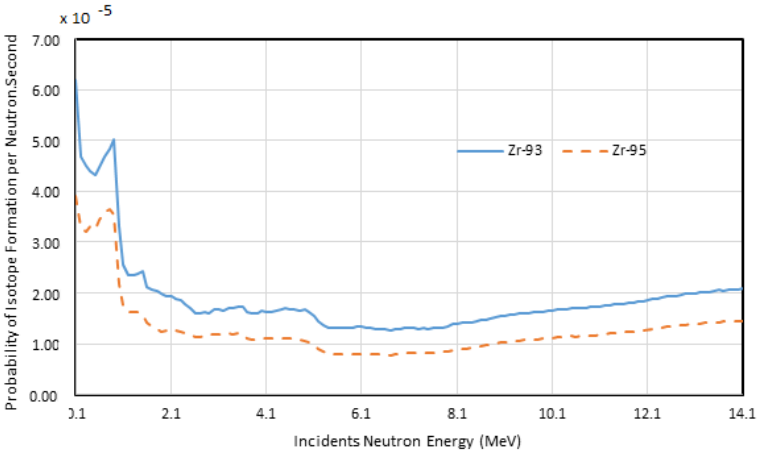

Fig. 2. Neutron interaction rates with Zr-92 and Zr-94 in Zirconium disulfide (ZrS_2).

The alpha particles emission probability is studied here to alternate the material's electronic structure due to their charged nature. Hence, we studied (n, alpha) interaction rates to report the mentioned isotopes' tolerance regarding alpha emission due to the neutron absorption. Fig. 2 shows the neutron interaction rate taken at different incident neutron energies with Zr-92 and Zr-94 for Zr-93 and Zr-95 production. Additionally, the probabilities of yielding Zr-93 and Zr-95 are at their highest at the epithermal energy range, as observed in Fig. 2. The neutron interaction rate with S-34 yields to S-35, as illustrated in Fig. 3. The probability of yielding S-35 is at its highest at the epithermal energy range. Hence, Zr-92, Zr-94, and S-34 can be considered transparent neutron isotopes. Therefore, they can form an excellent potential candidate for optoelectronic applications in nuclear power plants radiative environment. The obtained results agree well with the radiative capture cross-section of both Zr-92 and Zr-94, respectively. The comparison is shown in Fig. 4 (a), where the exact behaviour of the radiative capture cross-section of both Zr-92 and Zr-94 can be observed. Furthermore, the obtained results are in perfect agreement with the radiative capture cross-section of S-34, as shown in Fig. 4 (b).

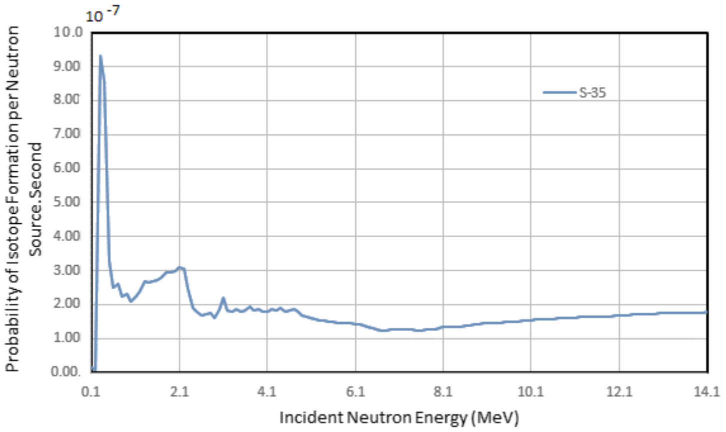

Fig. 3. Neutron interaction rates with Sulfur in Zirconium disulfide TMDC.

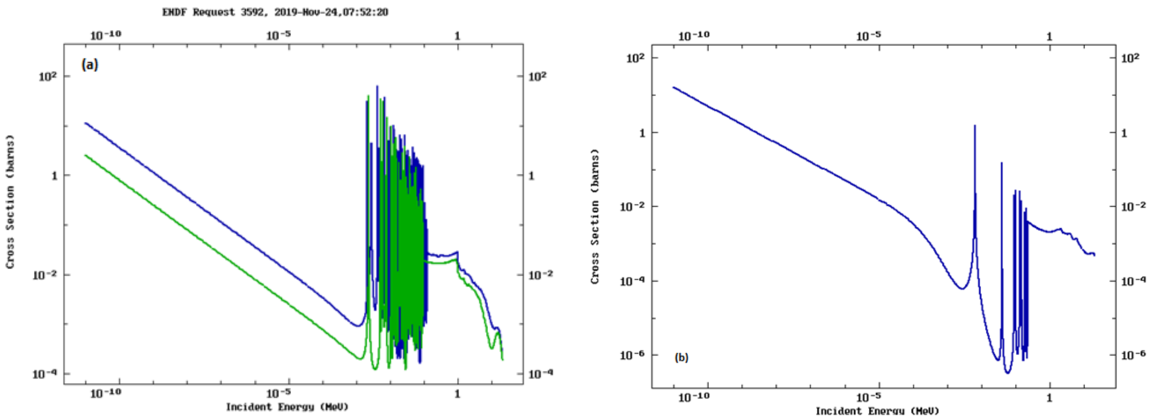

Fig. 4. Color online (a) the Zr-92 (in blue- the above curve) and Zr-94 (in green) Radiative Capture Cross-Section. (b) the S-34 Radiative Capture Cross-Section.

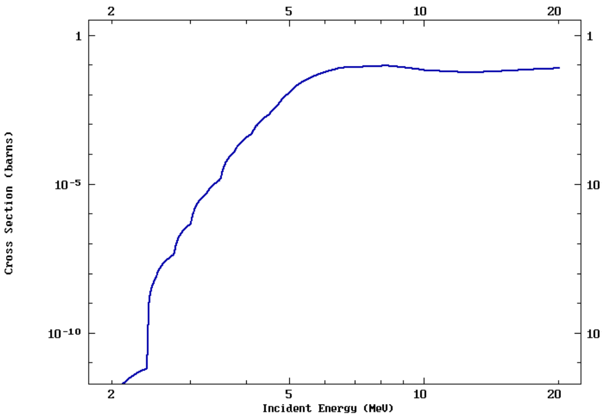

Fig. 5. S-34 n, alpha Cross-Section [10].

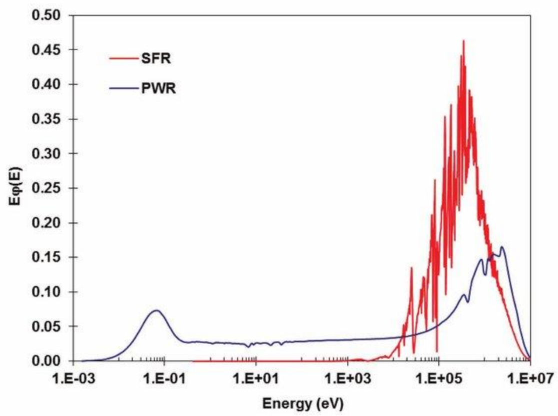

Fig. 6. The neutron energy spectra for operating PWR (thermal reactor) and SFR (fast reactor) from ref. [13].

The study is extended to the effect of the alpha emission due to neutron absorption. Fig. 5 shows the (n, alpha) interaction rates with S-34 in the Zirconium disulfide TMDCs. It can be noticed that there is a threshold for alpha emission due to neutron absorption (n, alpha) reaction. This threshold is at an energy value of about 2 MeV. Furthermore, the obtained results are found to be in a firm agreement with the (n, alpha) interaction cross-section behaviour, as shown in Fig. 5. The current work has been performed to study the effect of a nuclear reactor's radiative environment on ZrS_2. This study found that the highest neutron flux in a thermal nuclear reactor lies in the epithermal range of energy. Therefore, it has been found that alpha the range of epithermal range of neutron energies. Therefore, it can be concluded that the ZrS_2 TMDC material can be considered as a transparent neutron material regarding the alpha particle's emission due to neutron absorption. The obtained results introduce the ZrS_2 as a potential candidate that withstands being sent into space and used in optoelectronic devices such as photodetectors for monitoring applications in irradiative neutron environments the Fukushima reactor incident. The obtained results can be further discussed as follows. The neutron energy spectrum for a typical pressurized water reactor (PWR) and sodium fast reactor (SFR) is illustrated in Fig. 6. It can be observed that most of the neutron energies occur in the thermal and epithermal range for a PWR. Minimum transmutation will occur to ZrS_2 due to radiative capture and (n, alpha) interactions in an operating typical PWR environment. This reveals that ZrS_2 can be used in such a harsh environment. Therefore, it is considered as one of the potential candidates for optoelectronic applications inside an operating PWR.

On the other hand, for a fast reactor neutron energy spectrum, there is a high probability for (n, alpha) interactions as most neutron energy spectrum exceeds the alpha emission threshold (2 MeV). Since alpha particles are charged, this will affect the electronic properties of ZrS_2. Hence, ZrS_2 might not be a very good candidate to be subjected to a fast neutron radiative environment such as a fast reactor or a nuclear reactor meltdown accident such as the Fukushima accident.

4. Conclusions

In this work, we studied the radiative capture interaction rates for Zr-92, Zr-94, and S-34. Also, the (n, alpha) interaction rates for S-34 was studied. The purpose of this study was to check the ZrS_2 neutron radiative tolerance for photodetector monitoring applications in irradiative neutron environment such as nuclear reactors. The Zr-92, Zr-94, and S-34 are reported to be almost neutron transparent. The Zr-93 formation can be neglected from a material composition transmutation point of view due to its extended half-life. On the other hand, the Zr-95 and S-35 formation cannot be neglected as their yields have a half-life of 64.05 and 87.37 days, respectively. Further, the formed isotopes should be studied extensively as all of the decay in βmode will affect the electronic structure properties for ZrS_2 face perpendicularly to all wind directions and use only one pole, which can reduce a construction cost and enhance annual energy production.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

G. Laouini and M. Moustafa contributed equally to this work as first authors. G. Laouini contributed to the calculations; T. AlZoubi analyzed and validated the data; M. Moustafa wrote the manuscript and contributed to the visualization of the research, the analysis and validation of the results. All authors provided critical feedback and approved the final version.

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