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Investigation of epithermal and fast neutron shielding properties of Some High Entropy Alloys Containing Ti, Hf, Nb, and Zr

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Abstract

High entropy alloys often have excellent mechanical properties that conventional alloys based on one or two elemental combinations do not have. It is necessary to investigate whether these alloys can be used for nuclear applications with their properties such as high strength, fracture toughness, high corrosion and wear resistance. In this study, the thermal and fast neutron absorption properties of high entropy alloys with three different contents including Ti, Ta, Hf, Nb, and Zr elements were investigated. Their usability for nuclear applications has been demonstrated. In order to understand whether a material is neutron shielding, important neutron attenuation parameters such as effective removal cross section, half value layer, mean free path and neutron transmission factor (NTF) need to be determined. These reduction parameters were theoretically found with the Monte Carlo simulation GEANT4 code for epithermal and fast neutrons. It was found that Nb25 Ti25Hf25Ta25 has the best neutron shielding capacity among the investigated High entropy alloys. According to found all the results in the present work, we suggest that the all high entropy alloy samples can be used against any neutron leaks in nuclear operations.

Keywodrs High entropy alloys, Geant4, Neutron, Shielding

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1. Introduction

Radiation is widely used in energy production, medicine, archaeological, military, mining, space exploration, and investigations (WRIXON. 2013). Neutron radiation is widely used in condensed matter physics, crystallography, geology and mineralogy, biology, materials research, solid-state chemistry, and scattering and diffraction experiments. Neutron radiation has been successfully used in the destruction of tumors by at a high rate penetrating cells and tissues. During these processes, staff or patients can stay exposed to radiation. If exposed to high dose neutron, it can be damaged tissue, and cells such as vomiting, skin burns, acute ivegen, and the development of cancer. The best method of protecting against radiation is using quality shielding materials (PARK et al. 2014). Many samples were developed and produced for shielding neutron radiation such as stainless steel (AYGUN 2019a), (AYGUN et al. 2019b), (ALIM et al. 2022), (EID et al. 2022) some metals added alloys (RAMMAH et al. 2021), (EL-AGAWANY et al. 2021), (KORKUT et al. 2015) newly developed consist of metal oxide special glasses (AYGUN, et al. 2020a), (ELSAFI et al. 2021), (YIN et al. 2022) (AYGUN, et al. 2020b) reinforced heavy concretes (SARİYER & KÜÇER 2020), (AYGUN, et al. 2019c), (KINNO et al. 2002), (AYGUN et al. 2018) elastic and nonelastic high-density polymers (BILICI et al. 2021). (AYGUN et al. 2015), (HU et al. 2020).

High entropy alloys are a new type of alloy formed by using element concentrations from 5% to 35%. Incorporation into an alloy at the same mixing ratios in the same proportions ensures the formation of high entropy. High Entropy Alloys have properties such as high temperature, structural stability, strength, superior wear, and good corrosion resistance (MARY et al. 2015). High entropy alloys are excellent materials used in automotive, aerospace, gas turbine engine, exhaust nozzles, combustion chambers, and many similar applications. Due to their superior properties, these alloys are used as protective materials in nuclear applications like other sectors (PICKERING et al. 2021). Elastic composite materials containing low melting point polydimethylsiloxane and high entropy alloy are made for the radiation shield. GaInSnPbBi high entropy alloy was used as a filler to increase the radiation shielding ability and it was suggested that this alloy could be used for this type of composite structure. linear attenuation coefficient (LAC), tenth value layer (TVL), and effective atomic number (Zeff) were calculated using MCNPX software. It has been suggested that these composites can be used as a shielding material in X-ray applications (WANG et al. 2021). High entropy alloys (HEA) are preferred as a coating material in next-generation nuclear reactors III+ and IV because they have low thermal neutron cross sections and high melting points. HEAs were produced consisting of NbTiVZrx (x = 0.5, 1, 2) chemical formula according to it including Zr varied ratios experimentally and then cross-section calculated (KING et al. 2019). Low activation high entropy alloys (HEAs) with TiVZrTa and TiVCrTa chemical formulas were designed and produced for use as in-core, structural power reactors materials. After exposure to heavy ion implantation, the new alloys were found to have good irradiation resistance (KAREER et al. 2019). New developed TiTaHfNb, TiTaHfNbZr, and TiTaHfMoZr high entropy alloys are used in medical applications because of their superior resistance to corrosion and compatibility with human tissue (GUREL et al. 2021).

When a neutron with an energy of 1 MeV or more interacts with an alloy material, the neutron collides with an atom in the alloy and this atom gains a certain amount of kinetic energy and can be displaced in the molecular lattice region. This primary displacement causes the atoms and other atoms to be displaced by collision, causing the atoms to slide in the lattice. This event, which causes the displacement of a large number of atoms, causes the formation of spatial gap + interstitial Frenkel pairs. This is a primary damage to the material and can be permanent (ZINKLE, 2012). In fact, this movement causes an increasing temperature increase from the inside to the outside in the alloy, resulting in thermal structural deterioration. In the future, a new generation of fusion reactors will be built to ensure energy needs. In these reactors, high flux fast neutrons with 14.1 MeV energy will cause damage above100 dpa because they form hyper temperatures in the reactor components. This situation will cause high corrosion in protective components and can create cracks in the shielding material. Radiation leaks may occur as a result of this may occur at high entropy alloys can be used to prevent these leaks (KING et al. 2017).

In this presented study, the fast neutron radiation shielding parameters have been theoretically calculated for three high entropy alloys Monte Carlo Simulation Geant4 (GEometry ANd Tracking 4) software was used.

2. Theoretical background

A practical, convenient, and appropriate way to determine the severity of neutrons is to obtain the number of neutrons (N/cm^2) per unit area (n/cm^2) or flow rate (n/cm^{2}s). The flow of neutrons with an intensity I intensity is reduced by the shielding material with the x -thickness of the neutron source depending on the severity of the source and the coefficient of the neutron transmission (σnr). Similar to the Lambert-Beer Law used for the absorption of photons, the following statement for neutrons is valid:

$$I(x) = I_0 e^{-\Sigma_{nr} x} \tag{1}$$

Neutrons can interact with a material such as elastic, inelastic, neutron capture, or fission when colliding with it.

The probabilities of these interactions that neutrons can make with the materials they collide with are expressed by the effective removal cross-section and the total macroscopic cross-section (∑t), and this value can be calculated as follows. (AYGUN 2019 a).

$$\sum t = \sum N(\sigma t) \tag{1}$$

N; represents the number of nuclei per unit volume contained in the target material.

The total microscopic cross-section σt indicates the probability of interaction of neutrons of given energy with lower-density target materials, and this value is determined by the sum of the other scatterings, the microscopic sectional scattering (σs), and the microscopic section attenuation (σa).

$$\sigma_t = \sigma_s + \sigma_a \tag{2}$$

The effective removal cross-section, ∑R (cm2/g), refers to the probability of a fast or fission energy neutron's first collision with the shielding material it encounters. (SINGH & BADIGER 2014). If the neutrons interact with the shielding material in the form of a mixture, the effective removal sections for the mass-to-weight ratio of each element in this mixture can be determined as follows.

$$\sum R = \sum \left(\frac{\sum R}{\rho}\right) i \quad (3)$$

$\sum R$; is the effective removal cross-section of the shielding material and ρ; stands for density.
Half value layer (HVL) indicates the thickness of the material that halves the neutrons passing through the shielding material, and this value is found as follows.

$$HVL = \frac{ln2}{\sum R} \quad (4)$$

The mean free path (λ) shows the average distance that neutrons can travel in the target material they enter without first colliding, and this value can be calculated by the equation given to follow.

$$\lambda = 1/\sum R \quad (5)$$

The number of neutrons that pass through the shielding material with or without any interaction is determined by the Neutron transmission factor (NTF). The fact that this value is small indicates that the shielding material has high stopping power and this can be calculated as follows (SCHOBER, 2014), (SAHADATH et al., 2015).

$$NTF = \frac{I}{I_0} \quad (6)$$

I is the number of neutrons passing through the target material and I_0 is the number of neutrons coming into the material.

3. Material and Method

Geant4 Monte Carlo simulation technique

GEANT4 (Geometry and Tracing) is a Monte Carlo-based tool kit for predicting the events that particles and photons can generate when interacting with the target material. This kit is used in modern particle, high energy, and nuclear physics application experiments to predict situations where detectors and radiation can occur with target material at the same time. The Geant4 kit offers the opportunity to examine the interaction states of many elements and materials with radiation in a wide range of energy levels from eV to TeV' in hadronic and charged particles, electromagnetic and optical applications (AGOSTINELLI et al., 2003).
When using the program, all elements, molecules, compounds and material contents to be studied are defined in the detector and material interface library. At the start of the simulation process, this information is transferred to the worksheet. In this study, simulation calculations were performed for neutrons in the energy range of 3-14 MeV, using the GEANT4.10.2 version according to the geometry given in Figure 1.

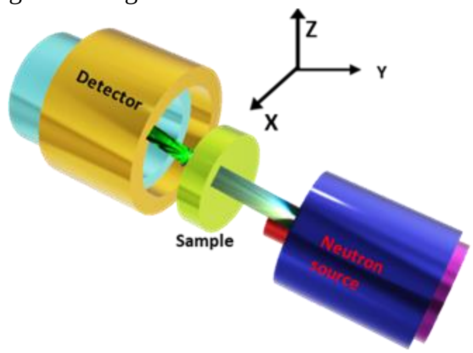

Figure1. Geant4 simulation geometry design

4. Results and Discussion

In this study, the samples (Ti25Ta25Hf25Nb25, Ti20Ta20Hf20Nb20Zr20, Ti20Ta20Hf20Mo20Zr20) whose chemical components are given in Table 1 were used. Some important neutron shielding parameters such as effective removal cross-section (cm^{-1}), half value layer (HVL), mean free path (MFP), and neutron transmission factor (NTF) were calculated with Geant4 code.

Neutron radiation attenuation properties

In this study, by using the Geant4 code for the simulation of neutron attenuation parameters such as removal cross-section, MFP, half value layer and transmission number were theoretically calculated with the simulation geometry in Figure 1.
The obtained data were compared with both, 316 LN stainless steel, which has (68.74Fe%+16%Cr+10%Ni+2% Mo+2% Mn+1%Si+0.15%N+0.045%P+0.03%C+ 0.03%S) chemical content and 8g/cm^3 density, and paraffin has ($C_nH_{2n}+_2$) chemical content and 1.2 g/cm^3 density.
The fast neutron attenuation parameters are given in Table 2 and Figure 2. of three types of high entropy alloys for 3-14 MeV energy.
The effective removal cross-section (cm^{-1}) is a useful parameter to consider in neutron shielding studies.
The greater the neutron effective removal cross-section value of a material to be selected as a shielding material, the greater the probability that the neutrons on the material will collide with the atoms of this shielding material. It is a desirable

property for a shielding material to be used for neutrons to have a highly effective removal cross-section.

The fact that this value is large indicates that the shielding material will also have a good absorption power. When Figure 2 is carefully examined, it is seen that the Effective Removal Cross-Section values of all studied samples decrease with increasing neutron energies. Because when the energies of the incoming neutrons increase, the elastic scattering numbers that neutrons can do with the material can also increase, and in this case, the possibility of interacting with the shielding material decreases. Looking at Figure 2, it is seen that the Effective removal cross-section values of all the samples examined are quite high compared to the paraffin selected as the reference sample, the HEA1 sample is higher than the 316 LN stainless steel and the others have close values. In particular, it was determined that the HEA1 sample had a higher value than all other samples. Considering these results, it is obvious that the shielding capacity of these HEA samples is high against fast neutrons.

Table 1 The chemical component of samples

Element	HEA1 (ρ=10.64 g/cm^3)	HEA2 (ρ=9.81 g/cm^3)	HEA3 (ρ=10.18 g/cm^3)
Ti	25	20	20
Ta	25	20	20
Hf	25	20	20
Nb	25	20	-
Zr	-	20	20
Mo	-	-	20

HEA (High entropy alloys)

Table 2
Comparison of high entropy alloys fast neutron shielding parameters for 2 cm sample thick and 10^5 incident neutrons

Sample code	Dose Energy (MeV)	Half Value Layer (cm)	Mean Free Path λ (mm)	Neutron Transmission Factor	Effective Removal cross section
P	3	15.098±0.150	21.7±0.212	0.63013	0.459
	6	22.282±0.221	32.1 ±0.312	0.73199	0.311
	9	22.720±0.227	40.0 ±4.157	0.77873	0.250
	12	28.999±0.289	41.8 ±0.286	0.78675	0.239
	14	32.383±0.323	46.7 ±0.480	0.80661	0.214
316LN	3	11.286±0.112	16.25 ±0.180	0.54082	0.614
	6	12.486±0.124	18.01 ±0.182	0.57361	0.555
	9	12.481±0.127	18.06±0.186	0.57402	0.515
	12	14.318±0.143	20.66±0.206	0.61597	0.484
	14	15.503±0.155	22.37±0.223	0.63953	0.447
HEA1	3	10.896±0.108	15.72 ±0.145	0.52898	0.636
	6	12.419±0.124	17.92±0.178	0.57185	0.558
	9	13.430±0.134	19.37±0.192	0.59665	0.516
	12	14.085±0.130	20.32±0.203	0.61098	0.492
	14	14.407±0.144	20.79±0.207	0.61769	0.481
HEA2	3	12.031±0.120	17.36±0.172	0.56160	0.576
	6	13.404±0.134	19.34±0.182	0.59619	0.517
	9	14.114±0.140	20.36±0.205	0.61144	0.491
	12	14.744±0.145	21.27±0.212	0.62478	0.470
	14	15.098±0.150	21.76±0.217	0.63160	0.459

HEA3	3	11.627±0.115	16.778±0.162	0.55068	0.596
	6	12.977±0.129	18.726±0.182	0.58579	0.534
	9	13.695±0.136	19.762±0.195	0.60246	0.506
	12	14.171±0.141	20.449±0.204	0.61317	0.489
	14	14.713±0.145	21.231±0.210	0.62390	0.471

Sample Code: P (Paraffin), **316LN** (Nuclear stainless steel), **HEA** (High entropy alloys)

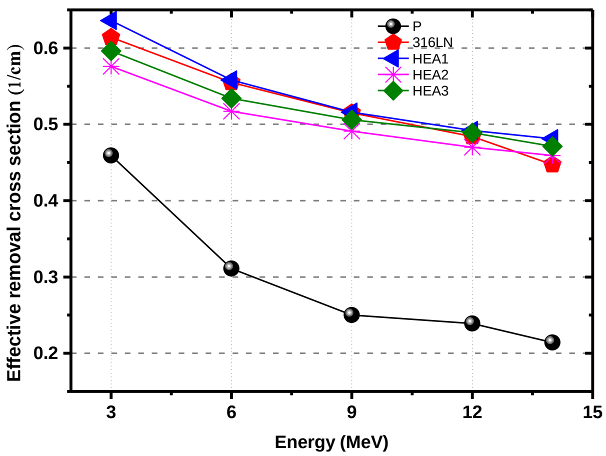

Figure 2. Theoretical 3-14 MeV Neutron Effective Removal Cross Sections

A material's low MFP values indicate that it has good neutron-stopping power (TEKIN et al., 2022). Looking at the values in Table 2' and Figure 3; the MFP values of all samples are lower than paraffin. Accordingly, all HEA samples have a greater neutron absorption power than paraffin. All these results showed that the ability of HEA samples to stop neutrons is high. Accordingly, all HEA samples have a greater neutron absorption power than paraffin. Again, while the MFP values of HEA1 sample are lower than 316LN stainless steel, these values of other samples are close. All these results showed that the ability of HEA samples to stop neutrons is high.

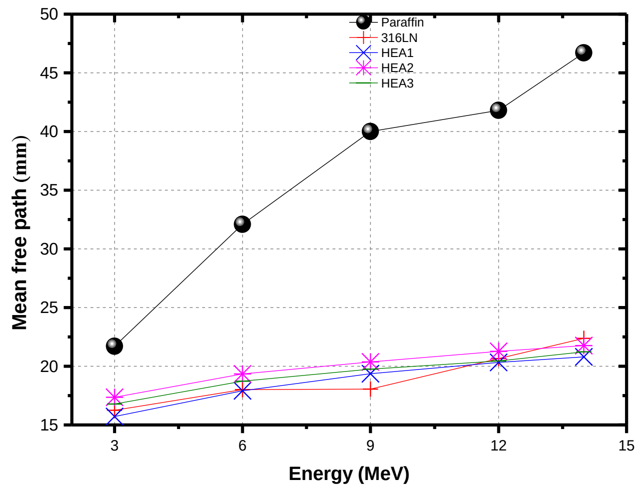

Figure 3. Variation of MFP values for different samples in the energy range of 3- 14 MeV.

Determining the number of neutrons that pass through the target material, which is particle-type radiation, is closely related to the power of that material to stop neutrons. Because the low number of neutrons passing through a material is proof that the material does not allow neutrons to pass through and as a result shows good shielding properties.

The number of neutron particles interacting with the shielding material is closely related to the neutron flux per unit volume. The amount of this flux depends on the probability of elastic, inelastic, scattering, and trapping interactions of neutrons with the shielding material. In other words, the determination of the neutron transition rate can provide information about the absorption capacity of shielding materials (SCHOBER 2014). It is desirable that this value is low.

In this study, transmission rates were determined by sending 10^5 neutrons to the materials. The results found are displayed in Table 2. According to Table 2, the transmission numbers of all HEA samples are lower than paraffin. Accordingly, the materials absorbed more neutrons than paraffin. Likewise, this value of the HEA1 sample is lower than that of 316LN stainless steel, so this sample has a higher rate of neutron suppression than 316LN. Other examples, HEA2 and HEA3, have values close to 316LN. They showed good stopping performance against neutrons in these samples.

Half value layer (HVL) refers to the material thickness that absorbs half of the neutrons interacting with the shielding material. A material to be used against neutrons should have low HVL values. Because neutrons will travel a long way in materials that are used too thickly, and during this time, they will cause the material to heat up by vibrating the material atoms as a result of their interactions with the atoms of the shielding material. Micro and macro deformations will occur due to thermal expansions in this heated shielding material, and as a result, the probability of neutron leaks will increase. The material half thicknesses, that is, the HVL values, which will halve the amount of neutrons incident on the samples, were calculated according to the increasing energy, and the changes of these values depending on the energy of the neutrons are given in Figure 4. When the figure is examined, the HVL values of all HEA samples are lower than paraffin. This result shows that all HEA samples have a higher absorption capacity than paraffin. The HVL value of the HEA1 sample is lower than that of 316LN stainless steel, which means that this sample has a superior absorption ability than 316 LN stainless steel.

HEA2 and HEA3 samples also have HVL values close to 316LN, and they showed good shielding properties.

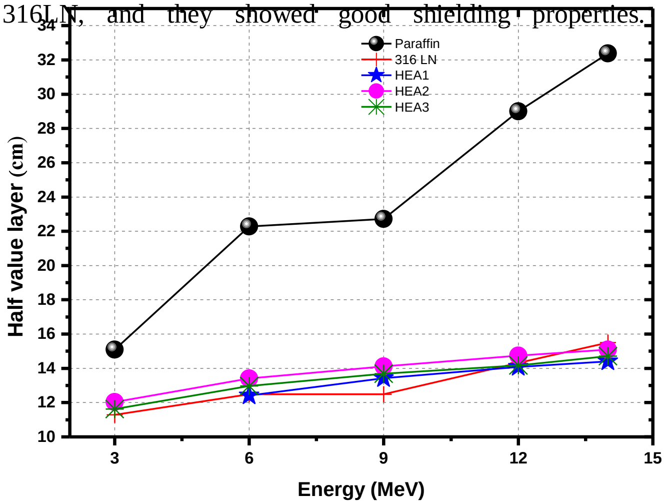

Figure 4. Change of HVL values in the energy range of 3-14 MeV

5. Conclusion

In this study, fast neutron absorption parameters of high entropy alloys with chemical content HEA1 (Ti25Ta25Hf25Nb25), HEA2 (Ti20Ta20Hf20Nb20Zr20), HEA3 (Ti20Ta20Hf20Mo20Zr20) were determined theoretically by Monte Carlo code. The shielding capacity of all examined HEA samples against neutron radiation was determined. It was observed that the neutron suppression power of all HEA samples was superior to the paraffin commonly used in shielding studies. It was determined that the neutron radiation stopping power of all examined HEA samples was higher than paraffin. In particular, the neutron radiation absorption capacity of the HEA1 sample was found to be greater than that of the reference sample, 316 LN nuclear stainless steel, and all other samples. However, it was observed that the absorption capacities of HEA2 and HEA3 samples were close to 316 LN samples. According to all results, it was determined that these HEA samples, which have excellent resistance to high temperatures as well as superior mechanical and structural properties, can be used as shielding materials to prevent neutron radiation leaks, especially in nuclear reactors, radiotherapy rooms, transportation and storage of used radioactive wastes, and it was suggested that they can be used safely in these applications.

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