Development of New Al-Ni-Cr-W Alloys for Enhanced Neutron Radiation Protection

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Abstract

Neutron radiation is utilized in many applications such as nuclear therapy, nuclear power plants, material analysis, space research, and more. Neutron leaks can occur in these applications, posing hazards to staff, operators, and therapy patients. Therefore, effective neutron shielding materials are always needed. In this study, two new types of neutron shielding alloy materials were developed, consisting of aluminum, nickel, chromium, tungsten, boron carbide, manganese, molybdenum, and silicium. The chemical composition and weight ratios of the composites were determined using the Monte Carlo Simulation's GEANT4 code. Mixing and molding methods were employed in the production of the alloys. Important neutron shielding parameters, such as the effective removal cross-section, half-value layer, mean free path, and radiation protection efficiency, were theoretically determined using the GEANT4 code. Additionally, fast neutron absorption capacities were measured using an Am-Be fast neutron source and a BF_3 portable neutron detector. The results were compared with 316 LN stainless steel. All new alloy samples were determined to have better fast neutron shielding capabilities than these reference samples. It was also observed that the new alloy samples exhibit both high-temperature resistance and mechanical durability. It is suggested that these new alloy samples can be used in neutron radiation shielding applications such as nuclear reactors, radioactive waste storage, and nuclear shelters.

Keywords: Neutron, alloy, geant4.

1. *Introduction*

Neutron radiation is a type of non-directly ionizing radiation released as a result of nuclear fission or fusion, and it can cause reactions in other atoms to produce new nuclides (Yue et al. 2013). Neutron radiation is commonly used in industry, diffraction and scattering experiments, material development and research applications, cosmology, oil and mineral research, and materials characterization studies. Neutron radiation does not ionize matter in the same way as electrons or protons, but when it interacts with matter, it can cause the release of ionizing radiation such as gamma rays. Neutrons do not have an electrical charge, so they can be more penetrating than gamma rays, alpha, or beta radiation, which makes shielding against them more difficult. Neutron radiation is used in Boron Neutron Capture Therapy to exterminate cancerous tissue. However, if adequate precautions are not taken, it can be hazardous to both personnel and patients. Neutrons may cause more DNA damage than other types of radiation due to their stronger interaction

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properties with tissues (Nouraddini-Shahabadi et al. 2024). Effective new shielding materials are always needed to protect against neutron hazards. In this regard, many new shielding materials have been developed, such as metal oxide-added glasses (Kaewkhao et al.2018), (Ekinci et al. 2024), (Alajerami et al. 2024), high-density strength alloys (Aygün et al. 2022), (Misned, et al. 2024),

stainless steels (Qi et al. 2022), (Oh et al. 2024), various chemical or organic molecules (Alaylar et al. 2021), (Aygün et al. 2020), (Aygün et al. 2024), and heavy concretes and bricks with added metals or oxides (Aygün, 2020), (Aygün and Karabulut 2018), (Makkiabadi, et al. 2024). Alloys are made by unifying two or more components with different characteristic properties. As a result, alloy materials can exhibit new properties such as high strength, temperature resistance, and corrosion resistance. Today, alloy and other composite materials are commonly used in various fields, including automotive, aerospace, aviation, military, healthcare, and construction. In recent years, alloys have also been used in nuclear technology due to their high durability. While alloy and composite materials often have high density, this property must be enhanced to withstand radiation effects (Sabhadiya, 2021).

Many alloy and composite samples were developed, and it is important to carefully consider the materials used, particularly regarding their neutron absorption capacity. Titanium (Ti) and Aluminum (Al) alloys are used in space research because they are lightweight, but they are not very durable against neutron radiation. To enhance their resistance to radiation, the radiation shielding capabilities of these alloys were improved using a vacuum plasma-sprayed method with hexagonal Boron Nitride (hBN) and titanium, incorporating 2-10 vol% of hBN. As a result, it was determined that the radiation absorption capacity increased by approximately 27% (Sukumaran et al. 2024).Fast and epithermal neutron shielding materials were produced for use with radioactive waste from nuclear reactors. The fast and epithermal neutron shielding performance was determined using the MCNP5 simulation program on an Al-Cd metal homogeneous mixture alloy. The results obtained showed that the neutron shielding performance increased by 10% in Cd-doped Al alloy samples (Heriyanto et al.2023). Shielding parameters were determined for neutron, electromagnetic, X-ray/gamma, and Bremsstrahlung radiations of Al–Li, Italma, Duralumin, Hiduminium, Magnalium, Hydronalium, Ni–Ti–Al, and Y-alloy. It was found that the Al–Ni–Ti alloy has good shielding performance compared to the other alloys against neutron, X-ray/γ radiation, Bremsstrahlung, and electromagnetic radiations (Sathish et al. 2023).

Polypropylene (PP)-based composites were developed, consisting of boron minerals such as ulexite, tincal, and colemanite. Fast neutron shielding parameters were calculated using Geant4 code, in addition to absorption experiments that were carried out. It was reported that samples with a higher content of colemanite have better shielding ability compared to other samples (Bilici et al. 2021). Epoxy resin-based composite samples were produced with the addition of lithium (LiF), chromium oxide (Cr_2O_3), and nickel oxide (NiO). To enhance temperature resistance, all samples were coated with sodium silicate paste. Neutron shielding parameters were determined both theoretically and experimentally. It was reported that these samples can be used for nuclear applications in fast neutron shielding studies (Aygün et al. 2020). Metal matrix composites have good mechanical and chemical strength, as well as high-temperature resistance; therefore, these materials can be used in nuclear technology. An Al-B_4C metal matrix composite sample was developed to determine its neutron shielding ability. It was reported that increasing the B_4C ratio in the composite leads to an increase in radiation absorption capacity (Gaylan et al. 2023).The radiation shielding capability of high entropy alloys (HEAs) such as CoNiFeCr, CoNiFeCrTi, and CoNiFeCrAl was calculated for gamma, electron, neutron, proton, and alpha radiations using the Phy-X/PSD, ESTAR (10 keV-20 MeV), and SRIM (10 keV-20 MeV) programs. It was determined that the CoNiFeCr alloy has better shielding performance than the other alloys (Sakar et al. 2023).

In this study, two new types of aluminum-based alloy samples were designed and fabricated. To evaluate their potential for nuclear applications, the neutron shielding capacities were determined through both theoretical and experimental measurements.

2. Neutron attenuation principles

Neutrons can interact differently with target materials through processes such as elastic or inelastic

scattering, absorption, capture, or complete stopping. The probabilities of these interactions can be described by the macroscopic cross section, effective removal cross section, half-value layer, mean free path, and radiation protection efficiency.
The macroscopic cross section can be calculated as follow

$$\sum = \frac{\rho}{A} N_A \quad (1)$$

the units of the quantity are in cm^{-1}.

$$N_A = \frac{\rho}{A} N_0 \quad (2)$$

N_A is the number of atoms of the absorption sample per (atom/cm^3), N_0 is the Avogadro's number (6.02.10^{23}), ρ is the density of the absorption sample, A is the molecular weight of attenuation sample.
The removal cross-section, ΣR, functions similarly to a macroscopic cross-section and can be utilized to assess neutron attenuation properties; however, it does not represent the probability of neutron-nucleus interactions. This parameter reflects interactions such as fast neutron energy loss, scattering, and capture, and its value is less than that of the macroscopic cross-section (El-Khayatt. 2010).This parameter is crucial for neutron protection applications. Additionally, it can be applied to composites, alloys, and mixtures, and it can be calculated using the following method.

$$\sum_R = \sum (\sum_{R/\rho})_i \quad (3)$$

$$\rho_i = w_i \rho \quad (4)$$

w_i is the weight percentage and ρ_i is the density of attenuation sample i.
When neutrons traverse a barrier material, they may lose half of their number or energy. The thickness of the material at this point is referred to as the Half Value Layer (HVL), which can be calculated using the following formula.

$$HVL = ln2/\sum R \quad (5)$$

The mean free path represents the average distance a neutron travels before colliding with atoms in the protective material, and it can be calculated as follows.

$$\lambda = {}^{1}/_{\sum_R} \quad (6)$$

Neutrons exhibit particle-like characteristics as a type of radiation, making the count of incoming or passing neutrons through a protective material essential in neutron shielding studies. To assess the shielding effectiveness of a sample, the neutron transmission factor needs to be calculated, which can be done as follows:

$$NTF = \frac{I}{I_0} \quad (7)$$

I denote the number of neutrons passing through the barrier sample, while I_0 signifies the number of neutrons incident on it. The neutron Radiation Protection Efficiency (RPE) offers valuable insights into the shielding capability of the material, and it can be calculated as follows (Sayyed et al., 2019).

$$RPE = 1 - \frac{N}{N_0}\, 100\% \quad (8)$$

Where N represents the dose that passes through the barrier material, and N_0 denotes the dose incident on the barrier material.

3. *Materials and Methods*

3.1. Monte Carlo simulation code GEANT4

GEometry ANd Tracking (GEANT4) is a Monte Carlo simulation framework utilized to assess the likelihood of radiation traversing various materials and the resultant interactions. This framework allows for the design of the geometries of radiation sources and materials, as well as the detection of secondary radiation and newly generated particles following the interaction of radiation with those materials. GEANT4 finds applications in high-energy and nuclear physics, medical physics, space exploration, military fields, agriculture, mining, and various other research domains, covering energy levels from eV to TeV. This toolkit facilitates the design of novel material shapes and the development of experimental models to study the impacts of radiation on both living and non-living entities. In this research, the toolkit was employed to create new alloys, choose chemical compositions for these alloys, and evaluate radiation shielding characteristics (Wellisch, 2005). Geant4 simülation geometry is given in Figure. 1.

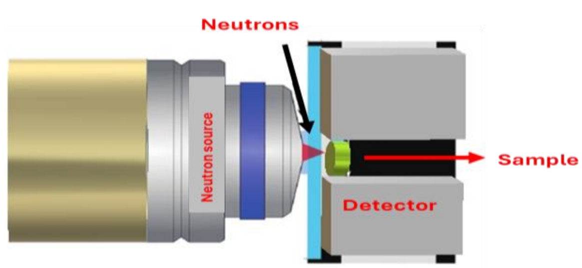

Figure. 1. Simulation geometry Geant4 3D visual

3.2. Sample preparation and experimental

Powdered forms of aluminum (Al), nickel (Ni), chromium (Cr), tungsten (W), boron carbide (B_4C), manganese (Mn), molybdenum (Mo), and silicium (Si) were blended uniformly for 20 minutes in a mixer, based on the composite ratios indicated by the Geant4 simulation results. Subsequently, this uniform mixture was subjected to cold pressing at 10 tons and 300 MPa to create pellets weighing 5g, with a thickness of 3 mm and a diameter of 1 cm, using the powder metallurgy technique. Each sample underwent tempering for one hour, where the temperature was gradually increased to 600 °C before cooling back to room temperature. The chemical compositions of these alloys are detailed in Table 1, and the produced sample images are presented in Figure. 2.

Table 1. Chemical composition ratios and density of alloy (AL) (%)

Material	AL1 (ρ=7.63g/cm^3)	AL2 (ρ=7.63g/cm^3)
Al	25	25
Ni	25	20
Cr	20	20
W	15	15
B_4C	15	-
Si	-	10
Mg	-	5
Mo	-	5

Al: Alloy

Figure. 2. Produced new alloy samples

In the dose measurement experiments, an Am-Be point fast neutron source and a BF_3 neutron detector were utilized. As shown in Figure 3, the experimental geometry was used for absorption measurements. First, the background dose (D0), which represents the dose emitted by the source, was determined. Then, each sample was placed between the source and the detector to be exposed to neutron radiation, allowing the absorbed dose measured by the detector (DD) to be recorded. Finally, the absorbed dose from the sample (DS) was calculated using the equation DS = D0 - DD.

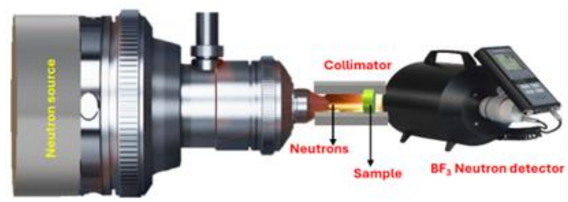

Figure 3. Neutron equivalent dose rate measurement system

4. Results and discussion

Aluminum alloys are both lightweight and exhibit good corrosion resistance and mechanical durability, making them preferable for nuclear applications (Sun et al. 2023). They provide effective shielding against low-energy radiation, such as α-particles and β-rays, but have limited shielding effectiveness against neutrons and gamma rays. To enhance their effectiveness for neutron shielding applications, aluminum alloys need to be reinforced with other metals. In this study, aluminum-based alloys containing various metals were designed and produced.

In Al-B_4C composites, the B_4C compound can be added to the alloy in amounts ranging from 5% to 50%. As the B_4C content increases in the alloy, the microhardness of the composite decreases, resulting in a reduction in its strength (Onaizi et al. 2024). To eliminate this disadvantage, the B_4C content has been kept constant at 15%. While studies generally focus on the shielding properties of Al alloys against thermal neutrons, this study investigates the shielding properties against fast neutrons (Jia et al. 2021).

4.1. Neutron absorption parameters

It is, of course, likely that there are differences between experimental measurements and simulation results. This is because in simulation studies, the sample is taken as completely homogeneous, and the experimental geometry is processed with exact dimensions. However, in experimental studies, it is not possible for the materials to be 100% homogeneous, and despite careful attention, some deviations in the geometry may still occur. In all studies, a margin of error of up to 10% between experimental measurements and simulation results is considered normal. However, if the shielding capacity of a material is well-predicted in simulation studies, good results are generally obtained in experimental measurements as well. In material design, the reactions of the materials to radiation can be determined in advance through simulation studies, allowing for the production of materials with the desired properties by using this prior knowledge in the manufacturing process (Kurt et al. 2020). In this study, keeping these considerations in mind, productions were made based on simulation results and experimentally verified.

The important neutron shielding parameters, such as effective removal cross-section, mean free path, half-value layer, and radiation protection efficiency, were theoretically calculated using the Geant4 code, and all results are presented in Table 2.

Table 2 Comparison shielding parameters in 5mm thick samples for 10^5 incident fast neutron (4.5 MeV)

Sample Code	Half value layer (cm)	Mean free path λ (cm)	Neutron transmission factor	Fast Neutron ERCS (cm^{-1})
316 LN	4.325	6.242	0.85194	0.1602
AL1	3.924	5.662	0.83828	0.1766
AL2	4.366	6.301	0.85321	0.1587

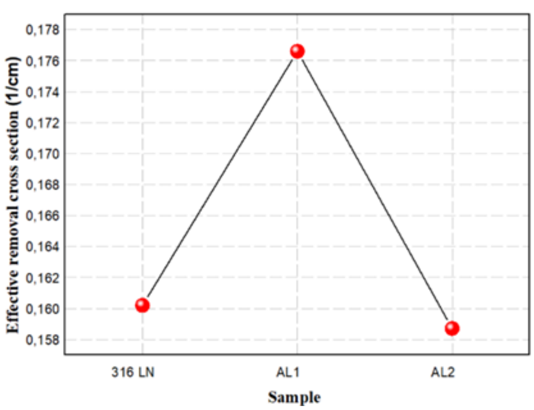

Figure. 3. Effective removal Cross Section (cm^{-1}) comparison of samples

When examining Table 2 and Figure 3, the AL1 sample has an effective removal cross-section value of 0.1766, while the AL2 sample has a value of 0.1587. In comparison, the 316 nuclear stainless steel has a value of 0.1602. According to these results, AL1 has a higher effective removal cross-section value than 316LN. A sample with a larger effective removal cross-section value indicates a higher shielding capacity (Manjunatha et al. 2019). Therefore, the AL1 sample has better shielding ability than the other samples. The AL2 sample also has good shielding capacity, but it is slightly lower than that of 316LN, although the difference between the two is small. If a sample has both a low mean free path value and a low transmission factor, it indicates a good shielding capacity for neutrons. According to Table 2, the AL1 sample has both a lower mean free path value and transmission factor compared to the reference sample 316LN. Therefore, the AL1 sample has better shielding performance than 316LN. The AL2 sample also has good shielding capacity, but it is lower than that of 316LN. Similarly, a low half-value layer (HVL) is a desirable property for shielding samples. It can be seen that the AL1 sample has a lower HVL value than 316LN, which indicates that AL1 has better shielding ability compared to 316LN. On the other hand, the AL2 sample has a higher HVL than 316LN, suggesting that AL2 has a lower shielding ability than 316LN. Based on all the theoretical results, the shielding abilities of the samples can be sorted as follows: AL1> 316LN>AL2.

4.2. Neutron absorption dose results

Table 3 gives experimental dose measurement results. As shown in Table 3, the incoming dose amount is 1.2987 µSv/h from the source. The AL1 sample has absorbed an amount of 0.5590 µSv/h, which is a ratio of 43.04%. Similarly, the AL2 sample has absorbed an amount of 0.5316 µSv/h at a ratio of 40.93%. The 316 LN sample has absorbed an amount of 0.5402 µSv/h at a ratio of 41.59%. According to these results, the AL1 sample has absorbed a greater dose than both the 316 LN stainless steel and the AL2 sample.

Al-3003 alloy was researched for its fast neutron shielding capacity, and it was determined that it has a shielding capacity of 3% (Samrah et al. 2024). However, the new alloy, Al1, has a shielding capacity of 44%, which indicates that this sample has excellent shielding ability.

However, the AL2 sample absorbed a lower dose than the 316 LN stainless steel, although the difference between the AL2 sample and the 316 LN sample is very small. These experimental measurement results indicate that all samples AL1>316 >AL2 have a relationship in terms of absorbed dose.

Table 3 Absorbed dose results of all samples

Sample	Absorbed Dose by Samples (µSv/h)	Radiation protection efficiency (%)
Background	1.2987	-
316 LN	0.5402	41.59
AL1	0.5590	43.04
AL2	0.5316	40.93

5. Conclusions

New types of alloys based on aluminum (Al) and nickel (Ni), with the addition of chromium (Cr), tungsten (W), boron carbide (B4C), manganese (Mn), molybdenum (Mo), and silicon (Si), were produced using the powder metallurgy method. The fast neutron shielding ability of these samples was determined both experimentally and theoretically, and the results were compared with those of 316LN stainless steel. According to the results, it is determined that both AL1 and AL2 samples exhibit good radiation shielding capacity.

It has been demonstrated that these new types of Al alloys can be effectively used against high-energy neutron leakage that may occur during the transportation and storage of radioactive waste and normal radioactive materials. Additionally, they can be used as protective shielding materials against fast neutrons in nuclear power plants, in boron neutron capture therapy applications in hospitals, and in military and space vehicles. Based on all these findings, it has been concluded that the newly designed and produced Al alloy samples are highly effective in radiation shielding and can be safely utilized in radiation protection applications.

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