Indian Journal of Pure & Applied Physics
Vol. 54, July 2016, pp. 443-448

An investigation on gamma and neutron shielding efficiency of lead-free compounds and alloys

Vishwanath P Singh[a,b]* & N M Badiger[a]

[a]Department of Physics, Karnatak University, Dharwad 580 003, India

[b]Health Physics Section, Kaiga Atomic Power Station-3 & 4, NPCIL, Karwar 581 400, India

Received 25 October 2013; revised 24 February 2014; accepted 11 July 2014

The effective atomic numbers, Z_{Pleff} and half value layer, HVL for lead-free shielding materials, tungsten compounds and alloys for gamma ray over an energy range from 1 keV to 100 GeV have been calculated in the present work. As well as the macroscopic effective removal cross-sections (Σ_R) for fast neutron (2-12 MeV) had been calculated. From results, it has been concluded that HVL and Σ_R values of the tungsten alloys are half of the lead shielding material. The values of Z_{Pleff} have been compared with possible experimental results, and good agreement has been observed. We found that the theoretical values of Z_{Pleff} of the tungsten alloys agree with experiment where the ratio of atomic number of the elements is near to unity and shows weak energy dependence for ratio away from unity. The tungsten alloys show better the degree of protection against gamma ray and neutron as compared to the lead, resulting in an overall reduction in thickness and volume of the shielding material. The tungsten alloys containing gold, silver, platinum, copper, nickel and iron were found superior lead-free radiation shielding materials. This study is expected to be useful for design and application of lead-free radiation shielding for nuclear engineering, radiation application to control lead hazard.

Keywords: Toxic, Shielding, Fusion, Lead-free, Protection, Weak dependency, Tungsten-gold

1 Introduction

Radiation protection is at the "cutting edge" of nuclear engineering and design where radiation exposure is controlled by various passive and active techniques. Conventional lead-based radiation shielding is widely used to offer protection against exposure to ionizing radiation because of its physical properties and ease casting, fabrication and malleability. It takes many forms; including lead impregnated rubber, glass, powder and lead-polyethylene-boron mixture for protection against gamma and neutron radiation and may form temporary or permanent arrangement of shielding.

Over the past years, a great deal of concern has been expressed about the toxicity of the lead[1-3]. The lead toxicity in children as well as adults has been studied and well documented. Corrosion in lead sheets of structural shielding walls has also been reported[4]. In view of this, an environment friendly non-toxic "lead-free" material is a challenging requirement for radiation shielding and protection. Various investigators have studied the gamma ray interaction parameters in silicate glass[5], industrial byproduct[6], tungsten-copper alloys[7], alloys[8,10-12] and carbon and stainless steels[13]. The effective atomic numbers of carbon and stainless steels[13] ascertain for computation of gamma ray interaction parameters for alloys.

As a result, we have studied gamma ray and neutron shielding efficiency of "lead-free" shielding materials, tungsten compounds and alloys (given in Table 1). Tungsten is being used for X- and gamma ray shielding, shielding glasses, in fusion reactor and around scintillation crystals[14-18]. In this work we have calculated the effective atomic numbers and half value layer of tungsten compounds and alloys for gamma ray over the energy range 1 keV to 100 GeV and macroscopic effective removal cross-sections for fast neutrons (2-12 MeV). The objective of the study is to determine the shielding efficiency of "lead-free" compounds and alloys, and comparison with lead shielding. This study should be useful for design of "lead-free" radiation shielding to control the lead hazard in nuclear industries.

2 Material and Method

Table 1 lists the tungsten compounds and alloys under present study. The mass attenuation coefficients,

*Corresponding author (E-mail: kudphyvps@rediffmail.com)

Table 1—Tungsten compounds and alloys for shielding materials

Compounds/Alloy	Chemical formula/Percentage composition
Tungsten oxides	WO_2, WO_3, W_2O_5, W_4O_{11}
Tungsten carbides	WC, W_2C
Lead Tungstate (PWO)	$PbWO_4$
Cadmium Tungstate (CWO)	$CdWO_4$
Tungsten-Copper	W50-Cu50, W55-Cu45, W60-Cu40, W65-Cu35, W70-Cu30, W75-Cu25, W80-Cu20, W85-Cu15, W90-Cu10 and W93-Cu7
Tungsten-Nickel-Iron	W85-Ni10.5-Fe4.5, W90-Ni7-Fe3, W90-Ni6-Fe4, W91-Ni6-Fe3, W92-Ni5-Fe3, W92.5-Ni5-Fe2.5, W93-Ni4-Fe3, W93-Ni4.9-Fe2.1, W93-Ni5-Fe2, W95-Ni3-Fe2, W95-Ni3.5-Fe1.5, W96-Ni3-Fe1, W97-Ni2-Fe1 and W98-Ni1-Fe1
Tungsten-Nickel-Copper	W90-Ni6-Cu4, W90-Ni5-Cu5, W90-Ni7-Cu3, W93-Ni4.9-Cu2.1, W93-Ni4-Cu3, W95-Ni3-Cu2, W95-Ni3.5-Cu1.5 and W96-Ni2-Cu2
Tungsten-Silver	W50-Ag50, W65-Ag35, W74-Ag26 and W90- Ag10
Tungsten Steel	W: 23, C: 0.77, Cr: 4.25, V: 1.6, Co: 11, Fe: 59.38
High Speed Steel	W:18, Fe:75, Cr:6, V:0.3, C:0.3
Tool Steel	W:17, Fe:66, Cr:10, C:3.2, Mo:2.5
Ferrotungsten	W:78, C:1, Fe;21
Ultimet	W: 2, Co: 54, Cr: 26, Ni:9, Mo:5, Fe:3, Mn:0.8, Si:0.08, C: 0.06
Hastelloy	W: 4, Co: 2.5, Cr: 16, Mo: 16, Fe: 5, Si: 0.08, Mn: 1, C: 0.01, V: 0.35, Ni: 55.06
Tungsten- Platinum	W15- Pt85 and W10- Pt90
Tungsten- Gold	W50-Au50
Tungsten-Nickel-Iron-Molybdenum	W: 93, Ni: 2, Fe: 2, Mo: 3

μ/ρ of the tungsten compounds and alloys have been computed by the mixture rule ($(\mu/\rho) = \sum_i^n w_i (\mu/\rho)_i$) where w_i is the proportion by weight and $(\mu/\rho)_i$ is mass attenuation coefficient of the i^{th} element taken from standard tables of XCOM program[19]. This calculates gamma ray interaction cross-sections of elements, compounds and mixtures at energies 1 keV to 100 GeV. The XCOM program is converted to windows platform is known as WinXcom[20].

2.1 Effective atomic numbers

The effective atomic numbers, Z_{Pleff} of the tungsten compounds and alloys can be calculated by various methods. In this work we have calculated Z_{Pleff} by total photon interactions formula[21] ($Z_{Pleff} = \sum_i f_i A_i (\frac{\mu}{\rho})_i / \sum_j f_j \frac{A_j}{Z_j} (\frac{\mu}{\rho})_j$), where f_i is molar fraction in the mixture or compound, μ/ρ is mass attenuation coefficient of i^{th} element, A is atomic weight, Z is atomic number and the ratio A/Z is approximately constant.

2.2 Half value layer

The half value layer, HVL is the thickness of the shielding material required to reduce the intensity of transmitted radiation to half of the incident upon it. The intensity is reduced by gamma interaction process, photoelectric absorption, Compton scattering and pair production. The HVL (HVL= ln2/μ) is derived using linear attenuation coefficients, μ. The μ is derived by multiplication of μ/ρ and the density of the material.

2.3 Macroscopic effective removal cross-section of fast neutron

The probability per unit length of a neutron losing all its energy above thermal is called the fast neutrons removal cross-section Σ_R (cm^{-1}). The effective removal cross-section for compounds and homogeneous mixtures may be calculated from the value Σ_R or Σ_R/ρ for various elements in the compounds or mixtures[22] by the general formula ($\sum_R = \sum_i \rho_i (\Sigma_R / \rho)_i$) where ρ_i and Σ_R / ρ are the partial density (the density as it appears in the mixture) and the mass removal cross-section of the i^{th} constituent, respectively. The Σ_R/ρ (in cm^2/g) values of elements have been taken from Kaplan and Chilton[23,24].

2.4 Errors

The errors in the present method for calculating μ/ρ is about 1% for low atomic number elements ($1 < Z < 8$) in the energy region 30 keV to 100 MeV[25]. Below 30 keV and above 100 MeV, the errors are as much as 5–10 %. For elements molybdenum ($Z > 42$) through uranium the error at low energies (10 keV to 1 MeV) ranges from 1–2 % from away from absorption edge to 5–10 % in the vicinity of absorption edge. Medical, biological and industrial, applications and transportation tend to use sources with photon energies above 5 keV. The values for effective neutron removal cross-sections calculated in the present work are accurate within 10% of the experimental values investigated for aluminum, beryllium, graphite, hydrogen, Iron, oxygen lead, boron carbide etc[26]. Therefore the errors in our results may not have any practical impact.

3 Results and Discussion

Values of the effective atomic numbers and half value layers of the tungsten compounds and alloys as a function of photon energy from 1 keV to 100 GeV are shown in Figs. 1 and 2, respectively. Table 2 shows the comparison of theoretical values with experiment results of some alloys. Table 3 provides the calculated fast neutron removal cross- section of tungsten compounds and alloys.

3.1 Effective atomic numbers

The variations of the effective atomic numbers, Z_{Pleff} of the selected tungsten compounds and alloys as a function of photon energy are shown in Fig. 1 (a-h). From the graphs it is evident that Z_{Pleff} varies with photon energy, with a peak in the proximity of the *K* shell absorption edges of the high atomic number elements, a minima at inter-mediate energy ($0.3 < E < 3$ MeV) and then becomes constant at high energy (>100 MeV). These variations are due to the different photon interaction mechanisms, namely photoelectric absorption, Compton scattering and pair production. The variation is large below 100 keV where photoelectric absorption dominates and above 3 MeV where pair production becomes the major interaction mechanism but varies little between 0.3 to 3 MeV where Compton scattering dominates. The alloys containing high atomic number elements show higher values of Z_{Pleff} as seen in Fig. 1 (h). The highest value of Z_{Pleff} across the entire range of energies for the lead (containing low antimony alloy) (Pb: 96.69 and Sb: 3.1) was also plotted for comparison of Z_{Pleff} (Fig. 1(h)).

The variation of Z_{Pleff} with energy can be explained by the dependence of the total interaction cross-section on atomic number and photon energy. This is proportional to $Z^{4-5}/E^{3.5}$ for photoelectric absorption, Z/E for Compton scattering and Z^2 for pair production. The Z_{Pleff} for oxides and carbide of tungsten were found lowest in Compton scattering region (Fig. 1 (a-b)). The Z_{Pleff} of lead tungstate was found higher than cadmium tungstate. The tungsten carbides are superior gamma ray shielding than tungsten oxides (Fig. 1 (a-b)). Fig. 1 (c, d, e and f) shows identical behavior as a function of photon energy with the absolute magnitude of Z_{Pleff} increasing with theweight fraction of tungsten in the alloy. If the

Table 2—Comparison of theoretical values and experimental results of effective atomic numbers of lead and tungsten-copper alloy at different energies

Energy (keV)	Effective atomic numbers						
	Lead		W65-Cu35			W60-Cu40	
	Exp.[a]	Present Work	Energy (keV)	Exp.[b]	Present Work	Exp[b]	Present Work
81	81.66	80.74	121.8	59.00	71.65	56.90	71.13
356	80.23	81.54	244.7	58.10	69.02	55.90	67.99
511	80.20	81.41	344.3	56.40	65.06	54.10	63.43
661.1	80.23	81.32	778.9	51.70	61.51	49.30	59.49
835	79.90	81.25	867.4	51.10	61.10	48.70	59.05
1274	80.37	81.15	964	50.60	60.74	48.00	58.66
1332	79.88	81.15	1085.8	50.10	60.38	47.70	58.27
			1112.1	50.00	60.31	47.60	58.19
			1408	48.40	59.93	46.90	57.79

[a]Kateb *et al.*[9]; [b]Murty[7]

Table 3—Macroscopic fast neutron removal cross-section of tungsten compounds and alloys

Compounds/Alloy	Σ_R (cm^{-1})	Compounds/Alloy	Σ_R (cm^{-1})
Tungsten Oxide (W_4O_{11})	0.1803	High Speed Steel	0.1554
Tungsten Carbide (W_2C)	0.0832	Tool Steel	0.1546
Lead Tungstate (PWO)	0.1231	Ferrotungsten	0.1629
Cadmium Tungstate (CWO)	0.1357	Ultimet	0.1645
Tungsten-Copper	0.1932	Hastelloy	0.1521
Tungsten-Nickel-Iron	0.2024	Tungsten- Platinum	0.2092
Tungsten-Nickel-Copper	0.2093	Tungsten- Gold	0.2079
Tungsten-Silver	0.1823	Tungsten-Nickel-Iron- Molybdenum	0.1982
Tungsten Steel	0.2752	Lead	0.1186

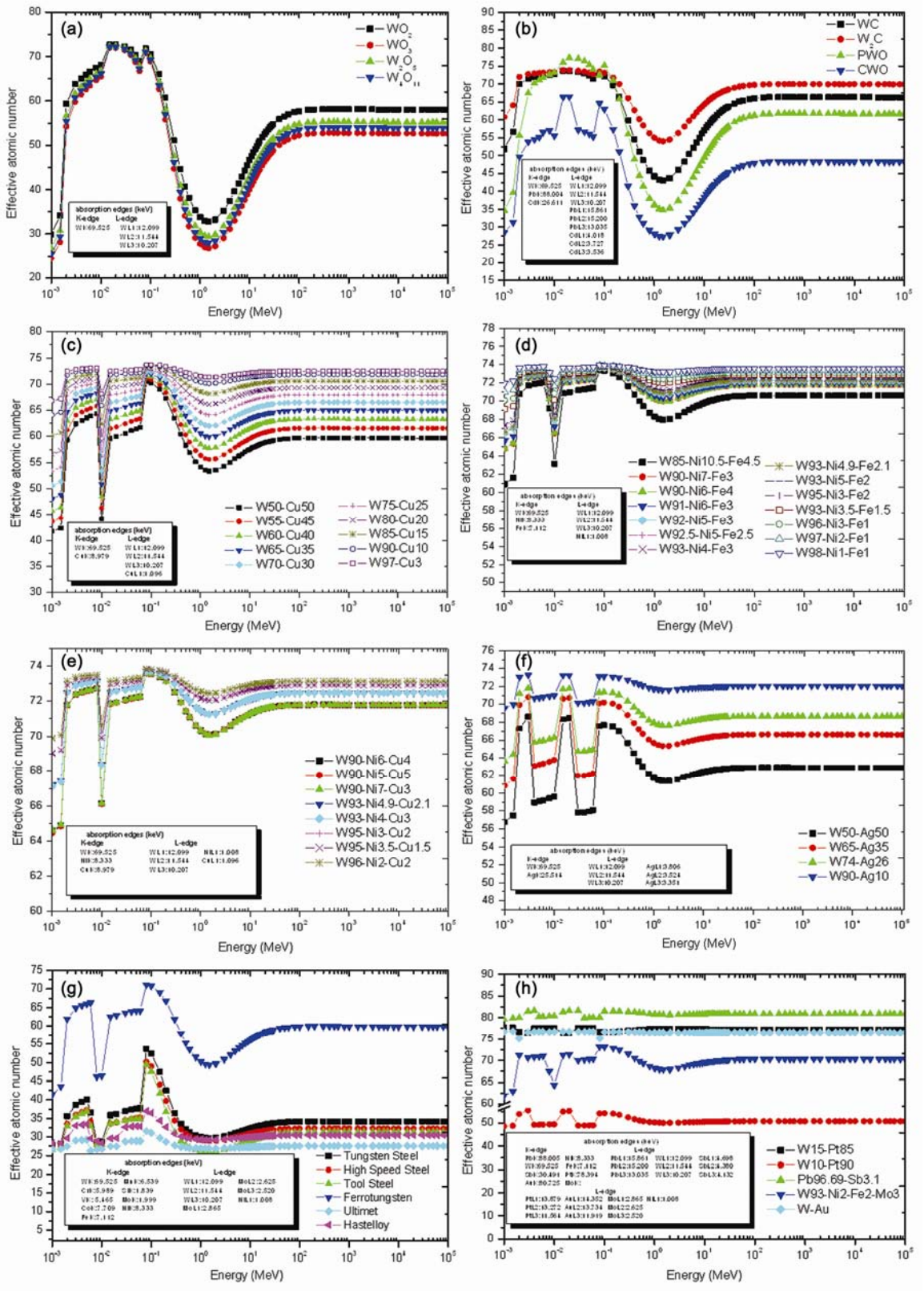

Fig. 1—The variation effective atomic numbers of tungsten compounds, alloys and lead in the photon energy ranging 1 keV to 100 GeV

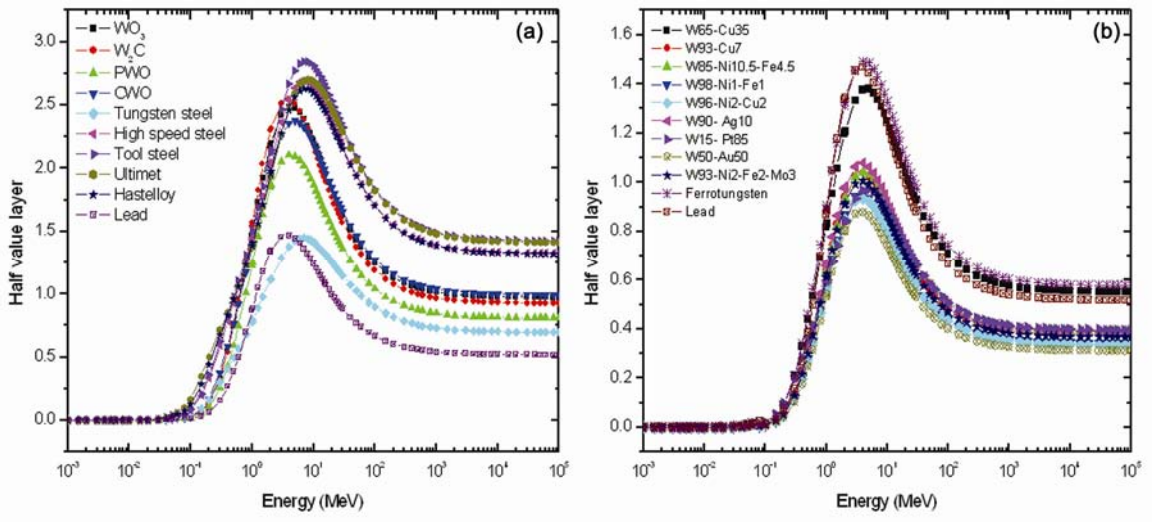

Fig. 2—The variation of half value layer of tungsten compounds, alloys and lead in the photon energy range 1 keV to 100 GeV

low atomic number elements are reduced, the alloys become less dependent on gamma ray energy higher energies.

In some alloys the variation of Z_{Pleff} with photon energy differs from compounds of the same heavy elements (Fig. 1 (h)). The presence of low atomic number elements in the compounds (*e.g.*, oxygen) makes a smaller contribution to the overall atomic cross-section, which consequently is less than that of an equivalent the alloy. The multiple peaks in the graphs in the low energy region dominated by photoelectric absorption are due to the *K* and *L* absorption edges of the elements in these compounds or alloys. The *K* and *L* absorption edges (in keV) are shown in the figures.

The Z_{Pleff} of the alloys of tungsten-platinum, tungsten-gold and lead (Fig. 1 (h)) show weak energy dependence in the energy regions dominated by photoelectric absorption and Compton scattering since they contain elements of similar atomic numbers. Small amounts of low atomic number elements change the Z_{Pleff} in the photoelectric absorption region but much less in the energy regions dominated by Compton scattering and pair production. However alcohols containing low atomic number elements (H, C and O) show a strong dependence on photon energy [27].

3.2 Half value layer

Figure 2 (a,b) shows the half value layer, HVL of tungsten compounds and alloys along with lead as a function of photon energy in range 1 keV to 100 GeV. The HVL for all the compounds and alloys considered here shows maxima in the photon energy region dominated by Compton scattering. It is evident that HVL of tungsten compounds and alloys containing significant amounts of low atomic number and low density elements (oxides, carbides, iron) is larger than for lead at all photon energies. These tungsten compounds and alloys would make inferior shields. Figure 2b show that the HVL as a function of photon energy for two element and multi-elements alloys containing a high proportion of tungsten and other high atomic number elements. With the exception of ferrotungsten, all have lower HVL's than lead and would make superior shielding. From Fig. 2 (b), it is to be noted that HVL values of W50-Au50 are the lowest. Therefore, the highest gamma ray shielding efficiency is provided by a 50-50 alloy of tungsten and gold though it may be expensive shielding.

3.3 Comparison with experimental results

This study of Z_{Pleff} of tungsten-copper and lead alloys has been compared with experimental results[7,9] and the results are listed in Table 2. Our theoretical estimate of the Z_{Pleff} of the tungsten-copper alloy slightly overestimates the experimental values, by as much as 25% in the energy region dominated by photoelectric absorption, but is in good agreement with experiment for the lead[9]. The experimental results do not show much dependence of the Z_{Pleff} on gamma ray energy. The slight disagreement with experimental values of the effective atomic numbers for tungsten-copper alloys may be due to the energy region selected falling between the influence of photoelectric absorption and Compton scattering, the

latter being much less dependent on atomic number. Also the atomic numbers of the lead (Pb: 82 and Sb: 51) are much closer in magnitude than for the tungsten-copper alloy (W: 74 and Cu: 29). It was observed that the theoretical Z_{Pleff} values of the tungsten alloys completely agree with experiment where ratio of atomic numbers of the elements is unity. The variation in the theoretical and experimental values of effective atomic number is due to non-unity of the ratio of atomic numbers consisting of one high- and one low-atomic number element.

3.4 Macroscopic fast neutron removal cross-section

The effective macroscopic fast neutron removal cross-section Σ_R (cm^{-1}) of the tungsten compounds and alloys is given in Table 3. It is highest for tungsten-steel. The Σ_R value of lead is very low (0.1186 cm^{-1}) as compared with tungsten alloys. The presence of gold or platinum in the tungsten alloy is superior to the presence of iron and these are at least twice as effective as lead as a shield against fast neutrons. Tungsten steel shows very high Σ_R values. The Σ_R values of the tungsten alloys are of order of stainless steel and copper alloys [28].

4 Conclusions

This work has shown that tungsten compounds and alloys are twice as effective neutron shields as lead shielding material. They are also better shields than lead against gamma ray over the photon energy range from 1 keV to 100 GeV since their half value layer is less. The effective atomic number of an alloy of tungsten with a low atomic number element is not greatly affected by the weight fraction of the lighter element.

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