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Gamma exposure buildup factors and neutron total cross section of ceramic hosts for high level radioactive wastes

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ABSTRACT

Mass attenuation coefficients and exposure buildup factors (EBF) for some ceramic hosts such as Hollandite ($BaAl_2Ti_6O_{16}$), Perovskite ($CaTiO_3$), Zirconolite ($CaZrTi_2O_7$), Apatite (Pb_{10} $(VO_4)_{4.8}(PO_4)_{1.2}I_2$), and Zircon ($ZrSiO_4$) for high level radioactive waste have been computed in the present paper. The mass attenuation coefficients for the Apatite were found to be the highest. The EBF for the Apatite were found the smallest in low-to-intermediate energy (<3 MeV). Neutron total macroscopic cross sections for the ceramic hosts were calculated for 2, 4.5 and 14.1 MeV using Geant4. Zircon for neutron low-energy (2 MeV) and Hollandite for high-energy (14.1 MeV) were found superior shielding materials. This study could be useful for radioactive waste management, handling, transportation, dose evaluation and other shielding requirements.

1. Introduction

Sources of nuclear radioactive waste are nuclear fuel cycle; mining and mining, reactor operation, spent fuel reprocessing, weapon operation, hospital, industries and nuclear research center. Nuclear reactors produce radioactive wastes in the form of long-lived radionuclides. Radioactive waste management is a vast and challenging area for effective utilization of fuel cycle. High level radioactive waste (HLW) arises from reactor operation contains high concentration of long-lived radioactivity. The compositions of HLW depend upon type of fuel, time of irradiation, neutron flux, cooling time, reprocessing chemistry etc. The list of fission products produced during reactor operation is given in Marples (1988). The HLW is being immobilized by vitrification, solidification, chemical or physical encapsulation. Immobilization is a technique to make easy in handling, transportation, storage and disposal of the HLW. Oxide glasses (borosilicate, phosphate, rare earth and sintered) and ceramics (synthetic rock, titanate, titania, zirconolite and phosphate) are reliable hosts for the HLW due to suitable properties of mechanical, thermal, chemical and radiation stability. The HLW contains different species of fission product (^{134}Cs, ^{137}Cs, ^{90}Sr, ^{144}Ce, ^{147}Pm, ^{151}Sm, ^{154}Eu, ^{155}Eu, ^{93}Zr, ^{99}Tc, ^{106}Ru, ^{107}Pd, ^{126}Sn) and actinides (Np, Pu, Am and Cm) (Marpel, 1988). The energy of gamma-ray emitted from the HLW ranges up to 3 MeV. The HLW in the form of spent nuclear fuel or vitrified reprocessing waste is highly radioactive and hot, requires heavy shielding and adequate cooling during its handling and temporary storage. Ceramics, glasses and concretes exhibit a very good shielding efficiency for gamma-ray and neutron radiations. Shielding properties for few glasses have been reported recently (Kaewkhao and Limsuwan, 2010; Chanthima and Kaewkhao, 2013). The gamma-ray and neutron shielding effectiveness of various concretes, glasses (Bashter, 1997; Singh et al., 2014), steels (Singh and Badiger, 2013a) and alloys (Singh and Badiger, 2014a) has been investigated. However, studies on shielding effectiveness of the ceramic hosts for the HLW are not found in the literature. This has encouraged us to study the shielding properties of the some ceramic hosts used for the HLW.

The intensity of gamma-ray beam through a medium follows Lambert's Beer law ($I = I_0e^{-\mu t}$) for narrow, mono-chromatic for thin absorbing material, where I and I_0 are transmitted and initial photon intensities, μ is linear attenuation coefficient and t is the thickness of medium. In case, any one of the conditions is not being met, this law is no longer applicable. The law is made applicable by introducing a correction factor called as "buildup factor, B". Now the modified equation is $I = B \times I_0e^{-\mu t}$ including the buildup factors, B. The B is 1 when all the above conditions are satisfied. The buildup is defined as the ratio of total value of a

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specified radiation quantity at any point to the contribution to that value from radiation reaching to the point without having undergone a collision.

The compilation for buildup factors by various codes is reported by American Nuclear Society (ANS, 1991). The data in the report covers energy range 0.015–15 MeV up to penetration depth of 40 mean free path (mfp). The buildup factors in the ANS report (ANS, 1991) are for 23 elements ($Z = 4$–92). Harima et al. (1986) developed a fitting formula, called Geometric Progression (G-P) which gives buildup factors of the good agreement with the ANS (ANS, 1991). Harima (1993) reviewed extensively and reported the current gamma-ray buildup factors. Various researchers investigated gamma-ray buildup factors for different materials; alloys (Singh and Badiger, 2014a), fly-ash brick materials (Singh and Badiger, 2014b), borosilicate glasses (Singh et al., 2014), brick materials (Singh and Badiger, 2013b), gaseous mixtures (Singh and Badiger, 2012) and human organs (Murut et al., 2011) which shows that the G-P fitting is a useful method for estimation of the buildup factors.

In view of above, mass attenuation coefficients and exposure buildup factors, EBF for some ceramic hosts like Perovskite ($CaTiO_3$), Zirconolite ($CaZrTi_2O_7$), Zircon ($ZrSiO_4$), Hollandite ($BaAl_2Ti_6O_{16}$) and Apatite ($Pb_{10}(VO_4)_{4.8}(PO_4)_{1.2}I_2$) have been computed first time. The EBF were calculated using G-P fitting method for photon energy range 0.015–15 MeV up to penetration depth 40 mfp. Neutron total macroscopic cross sections for the selected ceramic hosts were calculated at energies 2, 4.5 and 14.1 MeV using Geant4 Monte Carlo code. This study could be useful for radioactive waste management, handling, dose evaluation and other shielding requirements.

2. Computational methods

2.1. Mass attenuation coefficient

The mass attenuation coefficients, μ/ρ for the ceramic hosts were estimated using mixture rule as:

$$(\mu/\rho)_{\text{Ceramic}} = \sum_{i}^{n} w_i(\mu/\rho)_i, \tag{1}$$

where w_i is the proportion by weight and $(\mu/\rho)_i$ is mass attenuation coefficient of the ith element. The quantity w_i is given by $w_i = n_iA_i/\sum_j^n n_jA_j$ with condition $\sum_i^n w_i = 1$, where A_i is the atomic weight of the ith element and n_i is the number of formula units in the materials. The μ/ρ of individual element is taken from WinXcom program (Gerward et al., 2004). The linear attenuation coefficients for the ceramic materials can be estimated by multiplication of μ/ρ and density. Atomic mass and weight of the elements have been taken from recent IUPAC report (Michael et al., 2013).

2.2. Exposure buildup factors

The computational work for the EBF for the ceramic hosts is done in three steps as:

1. Calculation of the equivalent atomic number (Harima, 1983; Maron, 2007)
2. Calculation of the G-P fitting parameters (Harima, 1983; Maron, 2007)
3. Calculation of the exposure buildup factors (Harima et al., 1986; Harima, 1993)

The buildup of photons is mainly due to multiple scattering events by Compton scattering, so that equivalent atomic number, Z_{eq} is derived from the Compton scattering interaction process. The Z_{eq} for individual ceramic host is estimated by the ratio of $(\mu/\rho)_{\text{Compton}}/(\mu/\rho)_{\text{Total}}$, at a specific energy with the corresponding of an element at same energy using WinXcom program (Gerward et al., 2004). The Z_{eq} values of the ceramic hosts are calculated by logarithmic interpolation method (Harima, 1983; Maron, 2007) as:

$$Z_{eq} = \frac{Z_1(\log R_2 - \log R) + Z_2(\log R - \log R_1)}{\log R_2 - \log R_1}, \tag{2}$$

where Z_1 and Z_2 are the atomic numbers of elements corresponding to the ratios R_1 and R_2 respectively and R is the ratio for the ceramic host at a specific energy. The G-P fitting parameters are calculated in a similar fashion of logarithmic interpolation procedure for Z_{eq}.

The final step for buildup factor is putting G-P fitting parameters (b, c, a, X_K and d) in the equations given below:

$$B(E,x) = 1 + \frac{(b-1)(K^x - 1)}{K - 1} \quad \text{for } K \neq 1, \tag{3}$$

$$B(E,x) = 1 + (b-1)x \quad \text{for } K = 1, \tag{4}$$

where

$$K(E,x) = cx^a + d\frac{\tanh(x/X_K - 2) - \tanh(-2)}{1 - \tanh(-2)}, \tag{5}$$

for penetration depth $(x) \leq 40$ mfp

where x is the source-detector distance for the medium in terms of mfp and b, the value of the exposure buildup factor at 1 mfp, $K(E,x)$ is the dose multiplicative factor, and b, c, a, X_K and d are computed G-P fitting parameters which depend on the attenuating medium and source energy. The equivalent atomic numbers and G-P fitting parameters of the ceramic hosts are given in Tables 1–5.

Table 1
G-P fitting parameters for exposure buildup factors and equivalent atomic numbers for Perovskite.

Energy (MeV)	Perovskite					
	Z_{eq}	a	b	c	d	X_K
1.50E−02	17.92	0.135	1.009	0.487	−0.274	28.632
2.00E−02	18.09	0.255	1.027	0.315	−0.187	18.271
3.00E−02	18.26	0.242	1.077	0.355	−0.141	13.180
4.00E−02	18.36	0.218	1.169	0.388	−0.123	14.316
5.00E−02	18.43	0.208	1.294	0.422	−0.120	14.217
6.00E−02	18.48	0.173	1.425	0.496	−0.096	14.355
8.00E−02	18.54	0.121	1.686	0.630	−0.069	14.351
1.00E−01	18.59	0.070	1.892	0.780	−0.046	14.403
1.50E−01	18.65	0.008	2.184	1.017	−0.023	13.596
2.00E−01	18.69	−0.022	2.269	1.169	−0.004	11.956
3.00E−01	18.73	−0.044	2.265	1.290	−0.011	9.847
4.00E−01	18.75	−0.050	2.217	1.322	−0.010	8.717
5.00E−01	18.76	−0.064	2.127	1.365	0.015	21.250
6.00E−01	18.77	−0.064	2.074	1.357	0.013	18.396
8.00E−01	18.78	−0.062	1.982	1.331	0.014	16.814
1.00E+00	18.78	−0.057	1.916	1.296	0.015	15.874
1.50E+00	17.39	−0.043	1.819	1.212	0.013	16.223
2.00E+00	16.70	−0.031	1.757	1.148	0.008	15.173
3.00E+00	16.41	−0.008	1.659	1.057	−0.006	10.513
4.00E+00	16.35	0.009	1.586	0.997	−0.017	10.340
5.00E+00	16.31	0.013	1.515	0.979	−0.017	12.454
6.00E+00	16.30	0.020	1.467	0.958	−0.023	13.498
8.00E+00	16.28	0.031	1.387	0.929	−0.032	13.424
1.00E+01	16.26	0.036	1.324	0.919	−0.035	13.679
1.50E+01	16.25	0.051	1.230	0.891	−0.050	13.323

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