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Research paper

Investigation of clay bricks for storage facilities of radioactive-wastage

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ABSTRACT

Shielding is an integral part of any radiation facility, it is the method used for radiation protection. Safe storage of nuclear waste materials has become an important issue after Fukushima event. In Japan and France surface facilities are being considered for the storage of low-activity radioactive wastes. At these surface facilities, compacted clay liner obtained from clayey soil has been used as a cap barrier. The purpose of clay liner is to minimize the permeation of moisture, especially due to rainfall, through the barrier up to the radioactive material. Thereby it also reduces the emanation probability of undesirable radon gas and gamma-rays into the atmosphere. The brainchild behind the present investigation is to find the possibility of the burnt clay bricks to be used in these surface storage facilities. The present study aims to evaluate the gamma-ray shielding behaviour of burnt clay bricks in energy range 0.001–15 MeV. The samples of bricks were collected from four local brick-factories located near Bathinda-Ferozpur road, Punjab (India). An existing computer programme, Gamma Ray Interaction Coefficients (GRIC)-toolkit was modified for theoretical calculations of some interaction parameters for selected sample. Mineralogical study using X-Ray Diffraction (XRD) followed by Rietveld refinement (XRD-R) has been found in good agreement with Wavelength Dispersive X-Ray Fluorescence (WDXRF) investigation of samples for chemical compositions. XRD-R could be an interesting tool to recognize the radiation attenuation characteristics of materials. It has been concluded that local burnt clay bricks own the moderate gamma-ray shielding abilities while brick of clay (M-0) shows comparatively superior shielding behaviour. Thus brick of clay is the suitable candidate in the construction of environmentally-safe storage facilities for radioactive waste.

1. Introduction

Recent radiation leakage accident at Fukushima, Japan (2011) emphasized the awful need of systematic and precise studies for the durability and effectiveness of engineering materials used for the radiation shielding purpose. Consequently the safe storage of nuclear waste materials has become even a more important issue, universally. Surface facilities are being considered for the storage of low-activity radioactive wastes (Inui et al., 2012) and already have been used in France. Many such facilities dedicated to low and intermediate-level of nuclear wastes have been functional worldwide (Han et al., 1997). At surface facilities, compacted clay liner in combination with different soils and geosynthetic layers is generally used as a cap shield (Camp et al., 2010; Daniel, 1993). The purpose of clay liner is to limit the penetration of moisture into the waste (Rowe et al., 2004) and at the same time to restrain the possibility of release of radon gas to the environment. The clay liner is important not only in the storage of nuclear residues but also in landfills dedicated to hazardous or nonhazardous wastes (Koerner and Daniel, 1997). Using newly developed bending test along with X-Ray CT scanner for quantifying the deformation properties of compacted clay specimen, Mukunoki et al. (2014) showed that the cracks produced in the clay liner cause the fatal failure of the specimens to be used as shielding cover of the landfills.

Bricks produced from the mixture of clay and borogypsum (BG) have technological characteristics in agreement with the ASTM standards (C67-12, 2009). The addition of BG resulted in significant improvement in the compressive strength of the brick (Emrullahoglu Abi, 2014). On the other side common brick (burnt clay brick) shows the better gamma-ray shielding effectiveness than fly-ash and red-mud bricks (Mann et al., 2013). The present study aims to investigate gamma-ray shielding behaviours of burnt clay bricks and BG doped clay bricks and hence to verify their suitability to be used in safe nuclear waste storage facilities. Investigations of a composite material for its gamma-ray shielding behaviour require the evaluation of some standard interaction parameters such as; total mass attenuation coefficient (μ_m); Half Value Layer (HVL); Tenth Value Layer (TVL); analogous atomic numbers and

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Table 1
Chemical composition of selected brick samples (% by wt.).

Constituents	Clay burnt brick-samples				Doped clay bricks	
	CK-1	CK-2	CK-3	CK-4	M-0[a]	M-10[b]
Density (g cm^{-3})	1.73	1.74	1.75	1.73	1.82	1.71
% porosity	21.81	20.94	22.35	22.63	21.73	20.37
SiO_2	68.33	54.65	55.86	58.49	46.70	42.40
Al_2O_3	13.95	16.97	15.33	15.03	23.30	21.21
Fe_2O_3	4.89	5.53	5.37	4.90	8.13	7.40
CaO	4.14	5.88	4.71	4.41	3.89	6.33
MgO	2.13	3.02	2.93	2.68	2.17	2.13
MnO	0.08	0.11	0.11	0.10	0.00	0.00
Na_2O	1.90	2.71	2.63	2.40	0.81	0.73
K_2O	3.07	4.36	4.24	3.86	4.41	4.09
TiO_2	0.65	0.93	0.90	0.82	1.10	0.99
B_2O_3	0.00	0.00	0.00	0.00	0.00	0.42
P_2O_5	0.18	3.11	4.40	4.01	0.00	0.00
SrO	0.20	0.21	0.19	0.18	0.02	0.26
SO_3	0.39	2.40	3.21	3.02	0.14	3.28
L.O.I.[c]	0.09	0.13	0.12	0.11	8.96	10.43

[a] M-0; brick of clay (Emrullahoglu Abi, 2014).
[b] M-10; BG doped Brick (Emrullahoglu Abi, 2014).
[c] L.O.I.; loss on ignition.

effective electron density. These parameters have immense importance in deciding the effectiveness of the material for attenuating the intensity of gamma-rays.

2. Materials and methods

2.1. Clay brick materials

Four samples (CK-1, CK-2, CK-3 and CK-4) of burnt clay bricks were collected from four local brick-factories (with trade names: ND; BBC; RAJU and MGS) located near national highway (NH-15) (Bathinda-Ferozpur Road), Punjab (India). To make a broad comparison, two more clay brick samples M-0 (Brick clay) and M-10 (BG doped) based in Turkey were selected from literature (Emrullahoglu Abi, 2014). The sample M-10 is different from M-0 only in the way that it was modified by doping 10% of BG to the same clay due to which it attained the best compressive strength. On the other side all the local four samples (CK-1 to CK-4) are free from BG; however they are largely different from each other as well from M-0 and M-10 in oxide composition. The details of chemical composition of all samples have been listed in Table 1. The brick-samples were also tested for technological characteristics such as; water absorption, compressive strength according to ASTM C67 standards (2003). Wavelength Dispersive X-Ray Fluorescence (WDXRF) analysis was used for computing the oxides ratios and the results has been listed in Table 2.

2.2. Test methods for shielding behaviour

The test methods for the shielding effectiveness of a material in stopping (attenuating) gamma ray photons involve the computation and analysis of some standard gamma ray interaction parameters which are discussed as follows:

2.2.1. Porosity (ϕ) of bricks

Porosity (ϕ) represented by the ratio of the pore volume to the total volume of a sample. The change in ϕ directly affects the size, distribution and continuity of pores in the sample-brick (Hillel, 1998).

$$\phi(\%) = \left(1 - \frac{\rho_s}{\rho_p}\right) \times 100 \tag{1}$$

where, ρ_p (g cm^{-3}) and ρ_s (g cm^{-3}) are brick particle and bulk densities.

2.2.2. Total mass attenuation coefficient (μ_m) and atomic cross section (σ_a)

The methods to calculate the values of partial mass attenuation coefficient, total mass attenuation coefficient (μ_m) and Lambert–Beer law violation have been discussed in previous publications (Mann and Sidhu, 2012; Mann and Korkut, 2013).

The μ_m (cm^2 g^{-1}) value for composite material can be calculated as (Manohara et al., 2008):

$$\mu_m = \sum_i w_i (\mu_m)_i \tag{2}$$

where, w_i represents weight fraction and $(\mu_m)_i$ is total mass attenuation coefficient for i^{th} constituent element. The linear attenuation coefficient (μ, cm^{-1}) of the sample can be obtained by multiplying μ_m value with its density (ASTM D854-14, 2014). The atomic cross section (σ_a) can be obtained from the corresponding μ_m by the formula (Damla et al., 2011):

$$\sigma_a = \frac{\mu_m}{N \sum_i w_i / A_i} \,(\text{b atom}^{-1}) \tag{3}$$

where, N represents Avogadro's number, A_i is atomic weight of i^{th} constituent element of the sample.

2.2.3. HVL and TVL

Thickness of the absorber which reduces intensity of gamma ray to one-half (50%) of the incident value is termed as half-value layer (HVL). Similarly the thickness of the absorber which reduces gamma ray intensity to 10% of the incident intensity is termed as tenth value layer (TVL).

$$\text{HVL} = \frac{0.693}{\mu}\,\text{cm} \tag{4}$$

$$\text{TVL} = \frac{2.3026}{\mu}\,\text{cm} \tag{5}$$

2.2.4. Klein–Nishina cross section, ${}_e\sigma^{KN}$

The Compton total cross section (${}_e\sigma^{KN}$) obtained by Klein–Nishina (Davisson and Evans, 1952):

$${}_e\sigma^{eKN} = 2\Pi r_o \left\{ \frac{1+\alpha}{\alpha^2}\left[\frac{2(1+\alpha)}{1+2\alpha} - \frac{1}{\alpha}\ln(1+2\alpha)\right] + \frac{1}{2\alpha}\ln(1+2\alpha) - \frac{1+3\alpha}{(1+2\alpha)^2} \right\} \tag{6}$$

Table 2
Standardization of brick samples.

Symbols	Trade name	ASTM C618 (2003)			Ratio (Bormans, 2004)	
		$(SiO_2 + Al_2O_3 + Fe_2O_3) > 70$	$SO_3 < 3.00$	L.O.I. < 10.00	$(Fe_2O_3/Al_2O_3) > 0.33$	$(Fe_2O_3/CaO) > 0.80$
CK-1	ND	87.17	0.39	0.09	0.35	1.18
CK-2	BBC	77.15	2.40	0.13	0.33	0.94
CK-3	RAJU	76.55	**3.21**	0.12	0.35	1.14
CK-4	MGS	78.41	**3.02**	0.11	0.33	1.11
M-0	–	78.13	0.14	8.96	0.35	2.09
M-10	–	71.01	**3.28**	**10.43**	0.35	1.17