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# X-ray shielding behaviour of kaolin derived mullite-barites ceramic

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ABSTRACT

*Mullite-barite ceramic* (MBC) is an emergent material for effective shielding of redundant ionizing radiation exposure. The composition dependent mechanical, thermal, and microstructure properties of MBC that makes MBC a high performing novel radiation shielding candidate remained unexplored. This paper examines the possibility of exploiting Malaysian kaolin (AKIM-35) and barite ($BaSO_4$) derived ceramic (MBC) system for X-ray shielding operation. Using conventional pressing and sintering method six ceramic samples are prepared by mixing AKIM-35 with barite at varying contents (0, 10, 20, 30, 40 and 50 wt%). Synthesized pressed mixtures are calcined at 400 °C for 30 min and then sintered to 1300 °C for 120 min at a heating rate of 10 °C/min. Sintered samples are characterized via X-ray Diffraction (XRD), Field Emission Scanning Electron Microscope (FESEM), lead equivalent (LE), uniformity and dose reduction analyses. XRD pattern of prepared ceramics revealed the presence of monoclinic barium alumino-silicate (BAS) and orthorhombic mullite as major shielding phases together with other minor phase of barite and hexagonal quartz ($SiO_2$) structures. Furthermore, FESEM images of ceramics (between 0 and 30 wt%) displayed the existence of compacted monoclinic plate of BAS and acicular mullite morphology (ceramics at 40 and 50 wt%). Radiation tests displayed the capacity of ceramics (at 0 and 10 wt%) to shield the X-ray radiation emanated at tube potential range of 50–120 kV. The highest radiation attenuation is ascertained at 70 kV where the dose is reduced remarkably between 99.11% and 97.42%. Ceramics at 0 and 10 wt% demonstrated the highest lead (Pb) equivalent thickness (LE) of 0.44 mm and 0.34 mm, respectively. It is established that such MBC may contribute towards the development of shielding material against ionizing radiation in diagnostic radiology (X-ray) dose range.

## 1. Introduction

Over the years, ceramics and ceramic composites (CCs) have been used for several industrial and medical applications. In this regard, ceramic based structural materials that can withstand high temperatures (above 1200 °C) and can shield hazardous ionizing radiation dose are increasingly demanded. CCs are attractive due to their high melting point, low thermal expansion, strong oxidation resistance, environmentally friendly and low dielectric constant (Zhang et al., 1998; Amritphale et al., 2007a, 2007b). Barium alumino-silicate (BAS) based CC has served diversely as structural components in electronic packaging and biomedical applications for making cast prostheses owing to their physical opacity to ionizing radiation and higher mechanical strength compared to conventional feldspathic porcelains. Lately, BAS is used as a ceramic sealant in solid oxide fuel cell (SOFC) installed in the auxiliary power unit of vehicles or stationary power generator (Yang et al., 2003).

Few studies indicated that BAS can be used effectively as radiation shielding material. A radiopaque material using bauxite-red was developed by ceramic processing route. Shielding capability of red mud based shielding materials (RMSM) was compared with the conventional shielding materials such as concrete and lead. The half value thickness (HVT) of red mud based shielding materials in the energy range of 100–250 kV was found to be significantly lower than the conventional one. Meanwhile, BAS was prepared from fly ash (FA) to examine its X-ray radiation shielding ability (Amritphale et al., 2007a, 2007b). The FA obtained from power plants was mixed with barium compound to produce a new type of FA based radiopaque materials (FARM) following ceramic processing route with phosphate bonding. Systematic characterizations of FARM revealed that its HVT is considerably lower than concrete and lead in the aforementioned X-ray energy range. The XRD analyses confirmed the presence of monoclinic and hexagonal

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celsian together with sanbornite as the major shielding phases. Despite some studies a high performing BAS based composite such as *mullite-barite ceramic* (MBC) with optimum composition is far from being developed.

To fulfil such demand, for the first time we mix Malaysian kaolin AKIM-35 [$Al_2Si_2O_5(OH)_4$] with local barites ($BaSO_4$) to achieve improved shielding capability of mullite [$3Al_2O_32SiO_2$ or $2Al_2O_3SiO_2$]. Conventional pressing and sintering method is used to prepare MBC ceramics with different composition. As synthesized BAS based CCs (MBCs) are systematically characterized to determine their shielding efficacy in diagnostic radiology. The composition variation dependent structure, surface morphology and radiation shielding properties of MBC are inspected. The enhanced shielding capacity of MBC against X-rays is attributed to the presence of an optimum percentage of barium in the mullite. Results revealed that such MBC can be used directly to upgrade the normal room to the radiation room without altering the building structure. Furthermore, in case of flood or ablaze the proposed new material can maintain the integrity of the room and shielding capability without failure (Mann, 2017).

## 2. Materials and methods

### 2.1. Preparation of MBC composites

Well treated AKIM-35 and industrial grade of barium sulfate (barite powders) is used to prepare MBCs. These raw materials are sieved manually to remove the dust and sand present in the powder. The kaolin and barite powders are heated in an oven for 24 h at 105 °C to remove the water and moisture prior to the mixing process. Appropriate amount of AKIM-35 kaolin (100–50%) is mixed with barite (varied from 0 to 50 wt%) using a planetary mill Pulverisette-6 which employed 8 tungsten carbide balls to homogenize the mixture at 300 revolutions per minute (RPM) for 30 min. Conventional pressing and sintering methods are used to synthesize six ceramic green bodies. The detail compositions and the designation of the proposed MBC samples are summarized in Table 1.

A hydraulic press is used to press the mixture to achieve disc shaped samples of 4.0 cm diameter and thickness of 0.5 cm. The applied pressure is varied from 4 to 6 t to ensure the compactness of the disc shape structure MBC and devoid of cracks desired for the firing process. Sintering process is carried out in a Nabertherm HT16/18 electrical furnace at a heating rate of 10 °C/min. First, the calcination process is performed from ambient temperature to 400 °C for 30 min. Next, the sintering temperature is raised to a maximum of 1300 °C. During sintering, the mixture is subjected to soaking process for a period of 120 min at 1300 °C to ensure the optimum production of BAS from $Al_2O_3$ and barium (Choo et al., 2011).

### 2.2. Structural and morphological characterizations of MBC composites

The microstructure and morphological properties of the prepared MBCs are determined using X-ray diffraction (XRD) and Field Scanning Electron Microscopic (FESEM) imaging. XRD measurement is performed using a Bruker AXS D8 Advance Diffractometer (Germany) which employed Cu-$K_\alpha$ radiation. The post sintered phases (%), crystalline size and micro-strain are calculated using X'PertHighScore plus 2009 software (PANalytical). The surface morphology of MBC samples is imaged using ZEISS GeminiSEM 500.

**Table 1**
MBC sample's composition (wt%) and designation.

| Sample code | Kaolin (wt%) | Barite (wt%) |
|---|---|---|
| M1 | 50 | 50 |
| M2 | 60 | 40 |
| M3 | 70 | 30 |
| M4 | 80 | 20 |
| M5 | 90 | 10 |
| M6 | 100 | 0 |

### 2.3. Characterization for X-ray attenuation

Time ($t$), distance ($d$) and shielding ($s$) are the fundamental factors that control the exposure of external X-ray and γ-ray photons. However, for most of the practical applications (protection) shielding parameter is significant. Shielding is simply a barrier interposition between the source and the detector. The effectiveness of shielding for reducing exposure depends on the nature of the shielding (barrier) material and its thickness. The attenuated intensity $I$ (after transmitting) of an incident ionizing radiation beam of incident intensity $I_0$ through a shielding material over time ($t$) is given by:

$$I = I_0 e^{-\mu t} \quad (1)$$

where $\mu$ is the linear attenuation coefficient of the shielding material which is a function atomic number (Z) of the shielding material and the photon energy.

In the case of broad beam geometry, an additional term called build up factor (B*) exists due to scattering in the shielding material. Under this condition the modified equation takes the form:

$$I = B^* I_0 e^{-\mu t} \quad (2)$$

In the present study, the build-up factor is neglected due to the selection of narrow beam geometry and relatively shorter distance between the source and the detector. However, build up factor plays a significant role for larger source and longer distance. Materials with higher Z are preferred for gamma and X-ray shielding operations due to their good linear absorption coefficients. It is well known that gamma photons interact with matter via three major processes such as Photoelectric Effect (PE), Compton Scattering (CS) and Pair Production (PP), where μ is expressed as (Attix, 1986):

$$\mu = \mu_{PE} + \mu_{CS} + \mu_{PP} \quad (3)$$

Lead equivalent thickness (LE) is another important parameter, especially considered for radiation protection in medical applications. It is defined as the equivalent thickness of a shielding material corresponding to a given thickness of lead shielding. It must be emphasized that the LE of a material is a strong function of photon energy. At the higher energy range, only CS is significant and thus all materials absorb roughly the same amount of photons. Therefore, the difference between high and low Z materials is not that large. In the present work, the normalized dose rate ratios ($I/I_o$) for lead sheet thickness of 0.1, 0.2, 0.3, 0.4 and 0.5 mm are measured to produce a calibration curve for measuring lead equivalent thickness of other shielding materials.

Using standard procedures the uniformity test is conducted at five different positions on the disc shaped MBC sample to measure the attenuation distribution of X-ray beam of medical qualities (from KXO-50S machine operated with tube potential range of 50–120 kV). It is worth noting that to protect the on-site staff, patient and common public from harmful effects of ionizing radiation the thickness uniformity within the samples is kept less than 5%, which ensured the ultimate shielding of the proposed MBC samples.

## 3. Results and discussions

### 3.1. Physical properties of MBC samples

Fig. 1 shows the physical appearance of the synthesized MBC sample (M2) containing 40 wt% of barite. Sample M1 containing the maximum amount of barite (50%) revealed highest compactness upon sintering at 1300 °C. M1 appeared to be mostly cracked despite the movement of sintered samples to three different positions (inner, middle and outer part) inside the furnace. Sample M2 containing 40 wt% of barite is the