RESISTANCE OF BARIUM MULLITE CERAMIC AS RADIATION SHIELDING TOWARDS FIRE TEMPERATURE UP TO 1100 ˚C

Nur Syafika Suhaiman[1], Azuhar Ripin[2], Faizal Mohamed[1], Irman Abdul Rahman[1], Mohd Idzat Idris[1,*]

[1]*Nuclear Science Program, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia*

[2]*Medical Physics Group, Malaysia Nuclear Agency, 43600 Kajang Selangor, Malaysia.*

**Corresponding author: idzat@ukm.edu.my*

ABSTRACT

Barium mullite ceramics (BMC) is a potential radiation shielding produced by mixing a local powder of kaolin and barite through a conventional processing method. This ceramic will be used to replace the commercial radiation shielding such as lead because it is heavy and expensive. In addition, the integrity of lead will be degraded at high temperature due to its low melting point of 327.5 ˚C. This study was carried out to observe the resistance of barium mullite ceramic as radiation shielding towards fire temperature up to 1100 ˚C. Three samples of BMC were heated at 400 ˚C, 900 ˚C and 1100 ˚C each as a simulation in the event of a real fire. One sample was used as a reference (0 ˚C). Lead equivalent thickness (LET) tests were carried out with BMC of 4.5 mm thickness with capability value for X-ray machine from 60 kV – 120 kV before and after fire simulation test. The X-ray diffraction (XRD) tests and scanning electron microscope (SEM) observations were performed to evaluate the crystallinity properties and microstructures before and after fire simulation test, respectively. Results from LET tests showed that 4.5 mm thick BMC were equivalent to the range of 0.26 mmPb - 0.38 mmPb after the fire simulation. Meanwhile, celsian peaks were observed in XRD results at all temperatures. Furthermore, SEM observations showed no significant change in single phase of BMC even though it was heated at 1100 ˚C due to the high melting point of 1760 ˚C. From this study, it can be concluded that BMC performance as radiation shielding after the event of fire up to 1100 ˚C is consistent and has the potential to replace lead at X-ray room facility.

ABSTRAK

Seramik barium mullite (BMC) adalah berpotensi sebagai perisai sinaran yang dihasilkan melalui pencampurkan serbuk kaolin dan barit tempatan melalui kaedah pemprosesan secara konvensional. Seramik ini akan digunakan untuk menggantikan perisai sinaran komersial seperti plumbum kerana ia adalah berat dan mahal. Di samping itu, ketahanan pemerisaian bagi plumbum adalah berkurang pada suhu tinggi kerana plumbum mempunyai takat lebur yang rendah iaitu 327.5 ˚C. Kajian ini dijalankan untuk menguji ketahanan seramik barium mullite sebagai perisai sinaran pada suhu kebakaran sehingga 1100 ˚C. Tiga sampel BMC dibakar pada suhu 400 ˚C, 900 ˚C dan 1100 ˚C setiap satunya bagi tujuan simulasi sekiranya berlaku kebakaran sebenar. Satu sampel digunakan sebagai rujukan

(0 ˚ C). Ujian ketebalan kesetaraan plumbum (LET) telah dijalankan dengan BMC berketebalan 4.5 mm dengan nilai keupayaan tiub mesin sinar-X dari 60 kV - 120 kV sebelum dan selepas ujian simulasi kebakaran dilakukan. Ujian pembelau sinar-X (XRD) dan mikroskop imbasan elektron (SEM) dilakukan untuk mengenalpasti sifat kehabluran dan struktur mikro seramik sebelum dan selepas ujian pembakaran. Keputusan daripada ujian LET menunjukkan BMC berketebalan 4.5 mm adalah bersamaan dengan julat 0.26 mmPb - 0.38 mmPb selepas ujian simulasi kebakaran. Sementara itu, keputusan XRD menunjukkan puncak yang terhasil adalah fasa tunggal celsian pada semua suhu. Tambahan pula, pemerhatian SEM tidak menunjukkan perubahan ketara dalam fasa tunggal BMC walaupun ia dipanaskan pada suhu 1100 ˚ C berikutan takat lebur yang tinggi iaitu 1760 ˚ C. Daripada kajian ini, dapat disimpulkan bahawa prestasi BMC sebagai perisai sinaran selepas kejadian kebakaran sehingga 1100 ˚ C adalah konsisten dan berpotensi untuk menggantikan plumbum di kawasan bilik sinar-X.

Keywords: Barium mullites ceramic; Lead equivalent thickness; Fire simulation

INTRODUCTION

Radiation is an electromagnetic force comprising of particle flow or electromagnetic waves and magnetic energy that travels alongside the space at the speed of light [1]. Humans are exposed to radiation from the environment. Radiation will have effects in the short or long term if it is exposed to bodies. Therefore, the use of radiation shielding is necessary to prevent the radiation dose from human-made sources but could not prevent the radiation dose from natural sources. Commonly used radiation protection shields are lead-free materials. However, lead is high density and unsuitable for use at high temperatures because it has a low melting point of 327.5 ˚ C [2].

Ceramic such as barite has a potential for shielding. It was proved, to enhance the radiation properties, barite has been coated on fabric and is determined using a computer program written in C# language [3]. Furthermore, ceramics such as silicon carbide, aluminum nitrate, barium aluminum oxide and barium silicate have a high melting point, low thermal expansion, strong oxidation resistance, low dielectric heating and environmentally friendly that make it attractive to use as a radiation shielding and nuclear reactor components [4-7]. Barium mullite ceramic (BMC) is a candidate material to be used as a radiation shielding. This ceramic has a high heat resistance and excellent shielding elements [8]. Moreover, this ceramic is readily available because kaolin deposits in Malaysia are up to 112 million tons [9]. The BMC used in this study was fabricated from a mixture of kaolin and barite powder [10].

This research aims to evaluate the resistance of BMC as a radiation shielding at different temperatures up to 1100°C which can be applied in a real fire event by determining the value of lead equivalent thickness (LET) test. Changes in the microstructure of BMC samples before and after the fire test will be determined.

MATERIALS AND METHODS

Barium mullite ceramics

Kaolin and barite powder were dried using an electric oven to reduce moisture using a temperature of 105 ˚ C. The sample was then weighed according to 60% kaolin powder and 40% barite powder. They were mixed by using the Frits Pulvarisette-6 blender for 30 minutes at 300 rpm speed. Eight tungsten carbide balls were used to ensure that the powder was mixed smoothly.

The completed mix was divided into four samples. Each sample weights 13.25 g. Each sample was put in the mold for compression for 3 minutes using the 4.5 tons Hydraulic Model 3851-0 machine to produce disc-shaped samples with a thickness of 4.5 mm each. The samples were burned using the Nabertherm furnace with a temperature of 400°C for 37 minutes and then increased with a rate of 10°C/min up to 1300°C and hold for 90 minutes. Finally, samples were allowed to cool at room temperature overnight to produce a BMC [10].

Fire simulation test of barium mullite ceramic samples

A total of three samples marked with F2 - F4 undergo a fire simulation test [11]. F1 is a reference sample with a temperature of 0°C. F2, F3, and F4 were burned at 400 ˚ C, 900 ˚ C and 1100 ˚ C, respectively. All samples were burned using Muffle 1700 ˚ C furnace Model BSK-1700X-M for 4 hours per sample.

Lead equivalent thickness test with 4.5 mm thick ceramic

Lead and each BMC underwent the radiation attenuation test. The value of ln (I/I_o) that obtained from the lead test using an equation (1) were plotted to get a lead calibration curve. Then, lead equivalent thickness tests were performed using ln (I/I_o) of BMC value. All data collected were compared with the reference sample to evaluate the resistance of radiation shielding.

X-ray diffraction test and scanning electron microscope test

The X-ray diffraction test of each BMC at an angle of 10 ˚ - 80 ˚ using Bruker AXS D8 Advance was conducted to examine the crystallinity properties. Scanning electron microscope ZEISS Gemini SEM 500 was used to observe the microstructure of BMC. All changes were analyzed and compared with the reference sample.

RESULTS AND DISCUSSION

Lead equivalent thickness test with 4.5 mm thick ceramic samples

The radiation test for the lead sample was carried out to produce lead calibration curve using the following formula;

$$I = I_{\circ} e^{-\mu x} \tag{1}$$

The lead calibration curve in Figure 1 was plotted based on standard IEC61331-1 and was used to calculate the lead equivalent thickness with 4.5 mm thick BMC sample. The lead calibration curve graphs will be used as a reference for the LET test.

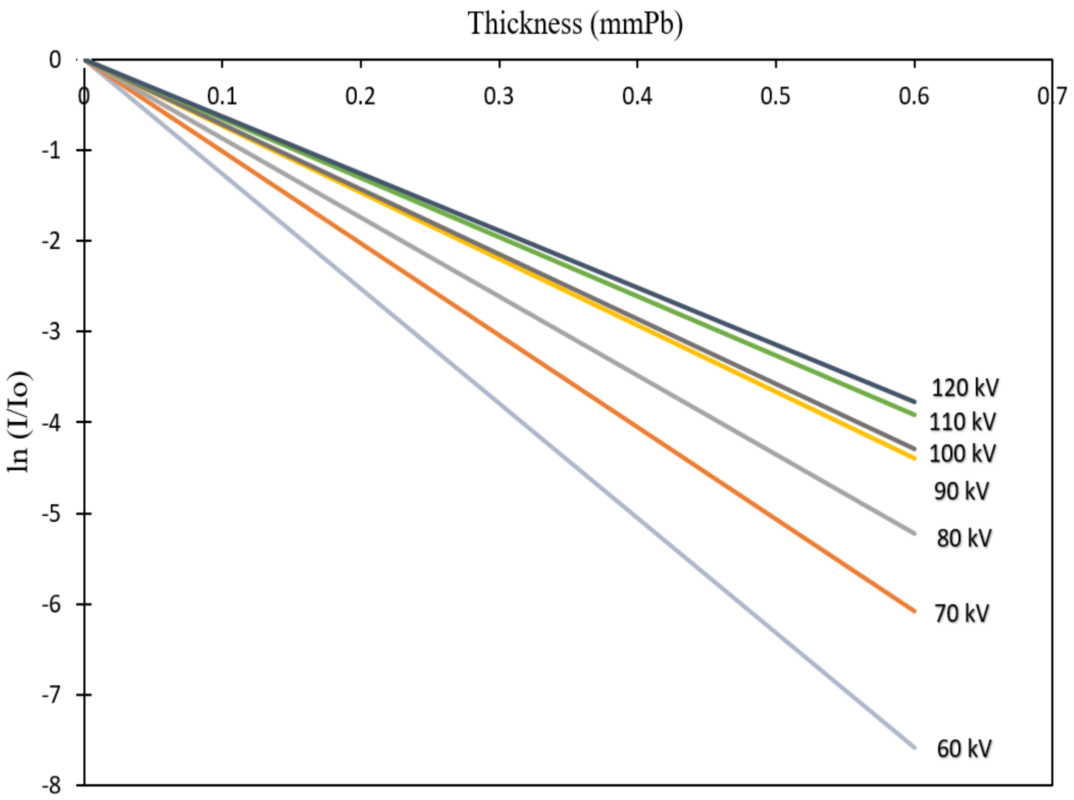

Figure 1. Lead calibration curve graphs (60 kV – 120 kV)

From the lead calibration curve, the range of LET test of BMC after the heat treatment was between 0.26 mmPb – 0.38 mmPb as shown in Table 1. However, there is no significant difference between the LET value before and after the fire simulation. It is learned that after heat treatment at 1250°C for 100 hours, no significant change of the microstructure was found in the Barium Aluminosilicate (BAS) glass-ceramics that prepared by hot-pressing BAS glass powder derived via the sol-gel method [12].

Table 1. Lead equivalent thickness with ceramic samples after a fire simulation

Samples	Lead equivalent thickness at different kV						
	60 kV	70 kV	80 kV	90 kV	100 kV	110 kV	120 kV
F1	0.26	0.32	0.33	0.37	0.34	0.36	0.35
F2	0.27	0.32	0.34	0.38	0.36	0.36	0.35
F3	0.26	0.31	0.34	0.37	0.35	0.36	0.35
F4	0.26	0.31	0.34	0.37	0.35	0.36	0.35

As an example based on Figure 1 and Table 1, the shielding ability of F2 for 90 kV is -2.78, which corresponds to the lead equivalent thickness of 0.38 mmPb. It can be concluded that 4.5 mm BMC is equivalent to nearly 0.40 mm lead in order to shield radiation of 90 kV X-ray. In Malaysia, the regulation that was implemented by the Ministry of Health for radiation facilities requires materials with a LET of 2.0 mmPb [13]. Therefore, this study showed that 22.5 mm BMC could be installed to replace the lead in radiation facilities.

X-ray diffraction test and scanning electron microscope test

Figure 2 shows the F1 – F4 sample analysis using XRD. From the results, each sample has the same diffraction spectrum pattern. All heated samples produce a single phase of celsian with a monoclinic form similar to the sample that did not undergo a fire simulation test [10].

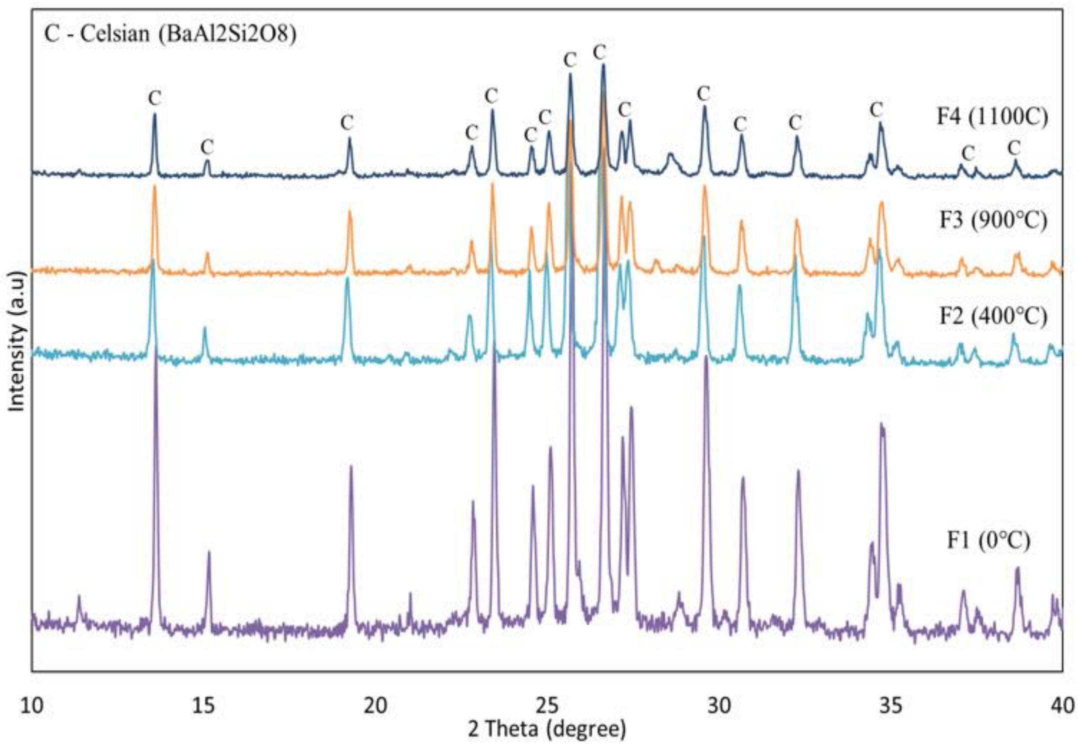

Figure 2. F1 – F4 samples analysis using XRD

The spectrum pattern was similar to that of post-fabricated BMC even though they were exposed to heat up to 1100 ˚ C, where the celsian with monoclinic phases remain the same and unaltered. It is because the celsian has a high melting point with a temperature of 1760 ˚ C [6]. Therefore, if a ceramic with a single phase of monoclinic celsian applied to a temperature below the melting point, no phase changes will occur [14].

However, it is clearly shown in Figure 2 that the crystallinity intensity of celsian peak decreased. It is known that the post-fabricated sample F1 has a high crystallinity phase [10]. In this study, as the BMC ceramics were annealed up to 1100 ˚ C, amorphization of the celsian phases seemed to occur. In contrast, XRD results for BMC which heat treated for 1 or 10 hours at various temperatures showed that the crystallization initiated at 1000°C and completed at 1100°C, where the hexacelsian to monoclinic celsian transformation was detected. Annealing treatments up to 1350°C continue to increase the relative amounts of celsian. However, the transformation is not complete even at the highest temperature [15].

Figure 3 shows the microstructure of samples heated at different temperatures observed by SEM with a grain size of 20 µm. Morphological images obtained through SEM supported the presence of celsian with a monoclinic phase for each F1 - F4 samples obtained through XRDSEM. All samples before and after the fire simulation test show that there is no significant change occurred.

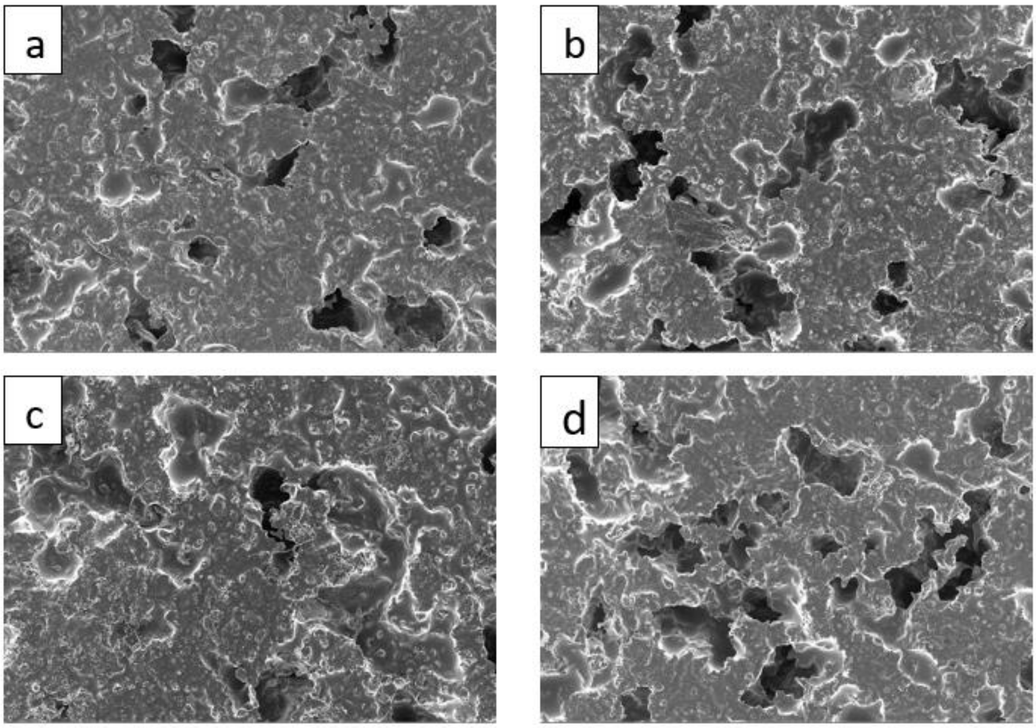

Figure 3. Microstructures of BCM heated at different temperatures using SEM.

a) F1 (0 ˚ C) b) F2 (400 ˚ C) c) F3 (900 ˚ C) and d) F4 (1100 ˚ C)

From the microstructure images obtained, however, it can be observed that the samples become more porous when they were heated to a high temperature up to 1100ºC. In a previous study using sillimanite ceramic, the porosity increased by 20 to 25% when the temperature was increased up to 1450 ˚ C [16]. Besides, thermal conductivity decreases when the number of porosity increases [17].

CONCLUSIONS

Results of the LET test with a 4.5 mm thick BMC were in the range of 0.26 mmPb - 0.38 mmPb after heat treatment. From the XRD results, it is found that the BMC produce a single phase of celsian with a monoclinic phase even after the fire simulations. SEM observations have shown that there is no significant change in the BMC microstructure. It can be concluded that the performance of BMC regarding radiation shielding is consistent in a fire event up to 1100 ˚ C. Therefore, BMC shows an excellent potential strength to replace lead material at X-ray room facility.

ACKNOWLEDGMENTS

The study was partly supported by a Geran Galakan Penyelidik Muda, National University of Malaysia, UKM (GGPM-2017-042). The experiments were performed with support from the staff of the Malaysian Nuclear Agency and Center for Research and Instrumentation (CRIM), UKM.

REFERENCES

[1] CNSC. 2012. Type and Sources of Radiation. Canadian Nuclear Safety Commission.

[2] Miru, M. 2014. Taburan Logam Arsenik (As), Kadmium (Cd), dan Plumbum (Pb) dalam Makrofit Akuatik Ipomea aquatica (Vol. 4, pp. 1–10).

[3] Akarslan, F., Molla, T., Uncu, I. S., Kilincarslan, S. 2015. Radiation Shielding Properties of Barite Coated Fabric by Computer Programme. SciTech Connect.

[4] Zhang, X.D., Sandhage, K.H., Fraser, H.L., 1998. Synthesis of BaAl2Si2O8 of Solid Ba-AlAl2O3-SiO2 Precursors: 11, TEM Analyses of Phase Evolution. J. Am. Ceram. Soc. 81 (11), 2983–2997.

[5] Amritphale, S.S., Anshul, A., Chandra, N., Ramakrishnan, N., 2007a. A Novel Process for Making Radiopaque Materials using Bauxite –Red Mud. J. Eur. Ceram. Soc. 27, 1945–1951.

[6] Long-gonzalez, D., Lopez-cuevas, J., Gutierrez-chavarría, C. A., & Pena, P. (n.d.). 2010. Synthesis of Monoclinic Celsian from Coal Fly Ash by Using a One-step Solid-state Reaction Process.

[7] MI Idris, H Konishi, M Imai, K Yoshida, T Yano. 2015. Neutron irradiation swelling of SiC and SiCf/SiC for advanced nuclear applications. Energy Procedia 71, 328-336

[8] Aral, N., Nergis, F. B., Candan, C. 2016. An alternative X-ray Shielding Material Based on Coated Textiles. Department of Textile Engineering, Istanbul Technical University, Turkey.

[9] Seong, K.P. 2005. Sustainable Mining of The Clay Resources in Peninsular Malaysia. Bull. Geol. Soc. of Malaysia 51: 1-5.

[10] Ripin, A., Mohamed, F., Choo, T. F., Yusof, M. R., Hashim, S., & Ghoshal, S. K. 2017. X-ray shielding Behaviour of Kaolin Derived Mullite-Barites Ceramic. Radiation Physics and Chemistry, 144. May 2017, 63–68

[11] ASTM E 119 Standard Test Methods for Fire Tests of Building Construction and Materials American Society for Testing and Materials, Philadelphia.

[12] Zhou W., Zhang L., Yang J. 1997. Preparation and Properties of Barium Aluminosilicate Glass-Ceramics. Journal of Materials Science 3 2 (1997) 4833 – 4836

[13] KKM, Division, E. S., & Of, M. 1995. Engineering Services Division Ministry of Health December 1995, 1984(December)

[14] Bansal, N. P., & Drummond, C. H. 1993. Kinetics of Hexacelsian to Celsian Phase Transformation in Journal of the American Ceramic Society, 76(5), 1321–1324.

[15] Mark J.H. and Narottam P.B. 1994. Crystallization Kinetics of Barium and Strontium Aluminosilicate Glasses of Feldspar Composition. NASA Technical Memorandum 106624. Ohio. NASA

[16] Boow J. and Walker G. E., 1946. Rate of Creep at Elevated Temperatures of Four Sillimanite Tank Blocks. J. SOC. Class Technol., 30 [137] 5-12T

[17] Zhang, H., Ma, S., & Wu, Y. 2011. Building Materials in Civil Engeenering. Oxford: Woodhead Publishing Limited.