Investigation of Radon Gas Emanation and Porosity Analysis of Organic and Carbon-Based Composite Brick

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Abstract. The key components in the production of bricks are pebbles, water and sand that emitted radon gas from the process of decay chain in Uranium-238. Application of cellulose nanofibrils (CNF) and graphene oxide (GO) have been broadly used for reinforcement materials due to their excellent physical, chemical and mechanical properties. However, the exploration of CNF, GO and the combination of both elements in cementitious materials to reduce radon haven't compressively studied. Alkaline treatment was implemented on pineapple leaves and sugarcane bagasse prior bleaching and high frequency ultrasonication. Graphene oxide was produced using modified Hummer's method. CNF and GO were combined via centrifugation to generate CNF/GO. Using Malaysian Standard (MS 7.6:1972), one control brick and composite bricks were made with different ratio of CNF, GO, and CNF/GO. Radon reading was recorded for 3 consecutive days using QuartaRad Radex MR107+ Radon Monitor in the airtight Perspex room. CNF was characterized using field emission scanning electron microscopy (FESEM). Surface porosity of the bricks was analyzed using Braunett-Emmet Teller (BET) analysis. Control brick has the highest radon value which is 67.1 Bq/m^3 and the lowest radon concentration was recorded in GO composite brick 1 with reading of 31.2 Bq/m^3.

Keywords: Cellulose nanofibrils, graphene oxide, nanocomposite brick, radon, radiation exposure.

INRODUCTION

Radon

Naturally occurring radioactive material (NORM) originated from radionuclide that can be found in the environment. The examples of the natural radionuclides also include primordial radioactive elements in the earth's crust and radionuclides from the cosmic radiation [1]. The most common elements are constituted from uranium, thorium, and potassium. Uranium (U-238) will release radium and radon gas along its decay series [2]. NORM is easily found in consumer goods like construction materials, granite countertops and phosphate fertilizers. Main components of the building material are from pebbles, soil, and stones that naturally emitted radon gas to the environment. Thus, natural radiation caused a great concern for the public. Radon was recognized as second factor to cause lung cancer after smoking [3]. Radon is a noble and inert gas that emitted from the Uranium-238 decay chain that is odorless, tasteless and colourless [2].

Nanomaterials have become promising substances in reinforcement materials due to their mechanical strength. However, there hasn't been much research done on the use of CNF, GO, or the combination of the two components in cementitious materials to lower radon concentration. This study investigated the radon levels of organic bricks constructed from pineapple leaf (PL), sugarcane bagasse (SCB) and carbon-based bricks made of graphene oxide (GO). The combination of both carbon and organic based materials in decreasing the radon levels have been explored in this study.

MATERIALS AND METHODS

Raw Materials

Sugarcane bagasse and pineapple leaf were obtained from the local place in Kota Bharu, Kelantan. Sugarcane bagasse fibre was retrieved after the extraction of sugarcane juice and pineapple leaf was scraped to get fibre. Graphite powder was supplied from Sigma Aldrich. 2.5 M NaOH and 0.4M Na_2So_3, 1000ml of distilled water was added separately to 99.9 g of sodium hydroxide (NaOH) and 50.4 g of sodium sulphate (Na_2So_3) pellet.

Preparation of CNF, GO and CNF/GO

Chemical and bleaching treatment followed with high frequency ultrasonication were applied to PL and SCB fibres. This process to eliminate hemicellulose and lignin in the fibre. During chemical treatment, fibres were immersed and boiled in NaOH and Na_2SO_3 for 8 to 12 hours. Then, it was rinsed off using hot distilled water 3 times. Bleaching treatment used hydrogen peroxide, H_2O_2 (2.5 M) and was kept for boiling around 6 to 8 hours until yellow-coloured fibres became white. It was rinsed again 3 times using cold distilled water and filtered using filter paper. After that, it was dried for 2-3 days as shown in Figure 2(b). After cellulose completely dried, 0.4 g of cellulose was added to 50 ml of distilled water. Cellulose was sonicated with high frequency, 20 kHz using QSonica500 for 30 minutes with an ice bath. CNF was produced.

Graphene oxide was synthesized from graphite using modified Hummer's method. 320 ml of H_2SO_4 was added to 80 ml of H_3PO_4. Then, 3 g of graphite powder and 18 g of $KMnO_4$ was added into the H_2SO_4 and H_3PO_4 solutions that was prepared before. The solution was stirred continuously using magnetic stirrer for 3 days as shown in Figure 3(a). After that, the solution was poured into iced distilled water and H_2O_2 was added as in Figure 3(b). It was washed in HCl for 10 minutes with 3 times repetition using centrifuge with 5,000 rpm. After that, it was centrifuged again in distilled water for 30 minutes until it turned dark brown, and the pH turned to 4.

CNF and GO were obtained from the previous steps and combined to form CNF/GO using Hettich Universal 32R centrifuge. CNF and GO were combined in a centrifuge tube with certain ratio and the mixture was centrifuged for 15 minutes at 5,000 rpm. Figure 1(a) displays inhomogeneous solution of GO and CNF before centrifugation process. The product of PLCNF, SCBCNF, GO, PL/GO and SCB/GO shown in Figure 1(b).

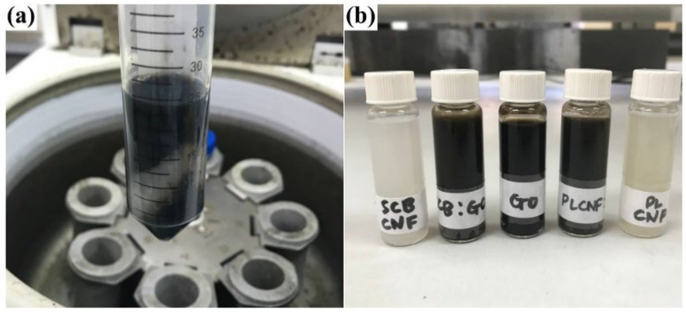

FIGURE 1. Image of (a) graphene oxide mixed with CNF before centrifugation process, (b) products of CNF, GO and CNF/GO.

Fabrication and Radon Monitoring of Composite Bricks

The bricks were fabricated using Malaysia Standard with measurement of 215 x 105.2 x 65 mm ± 7 mm. One control brick and composite bricks were fabricated using Malaysian Standard (MS 7.6:1972). Raw aggregates used were pebbles, sand and Portland cement and mixed by using the foaming agent and water. Four CNF composite bricks, GO composite bricks and CNF/GO composite bricks were fabricated with four ratios of CNF (ranging from 0.6 to 2.1% CNF), GO (ranging from 0.2 to 0.9% GO) and CNF/GO (ranging from 0.8 to 3.7% CNF/GO). QuartaRad Radex MR107+ radon monitor was used to measure radon gas emanation for 3 days as shown in Figure 2. The humidity was recorded using Govee Home Hygrometer.

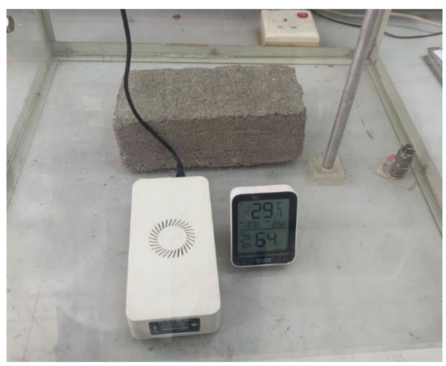

FIGURE 2. Image of radon monitoring with hygrometer.

Characterization of CNF and Bricks

Field Emissioning Scanning Electron Microscopy (FESEM)

The surface morphology of CNF and brick fracture has been observed using Quanta FEG 650 at Earth Material Characterization Lab. CNF was dropped onto the ITO glass and the brick fraction was taped onto the holder and both samples undergo gold sputtering process to increase the conductivity of the samples.

Braunett-Emett Teller (BET)

To study the surface porosity of the brick fracture, BET analysis was conducted. Brick fraction was degassed for 3 hours at 80˚C. the samples were analyzed using Micromeritics ASAP 2020-Accelerated Surface Area and Porosimetry Analysis at School of Chemical Sciences, Universiti

Sains Malaysia. The pore size was calculated using Alpha-S method and the formula as stated in Equation 1:

$$Pore\ Size = \frac{Q_0 V_{mol}}{22414 cm^3 STP} \tag{1}$$

The comparison of the pore sizes of the bricks was done for the highest and lowest radon concentration in all types of composite bricks.

RESULTS AND DISCUSSION

Surface Morphology of Composite Bricks

From FESEM images in Figure 3(a), control brick has cracks and produces big pores on the surface. In comparison, the pores are less visible in GO, PLCNF and SCBCNF composite bricks. As observed in Figure 3(b) GO composite brick has more uniform and compact surface compared to other composite bricks. Application of GO as nanomaterial substrate layers could strengthen the microstructure of hardened cement mortar by covering pores and cracks, thus making the mortar more homogeneous, compact, and preventing the development of microcracks [4].

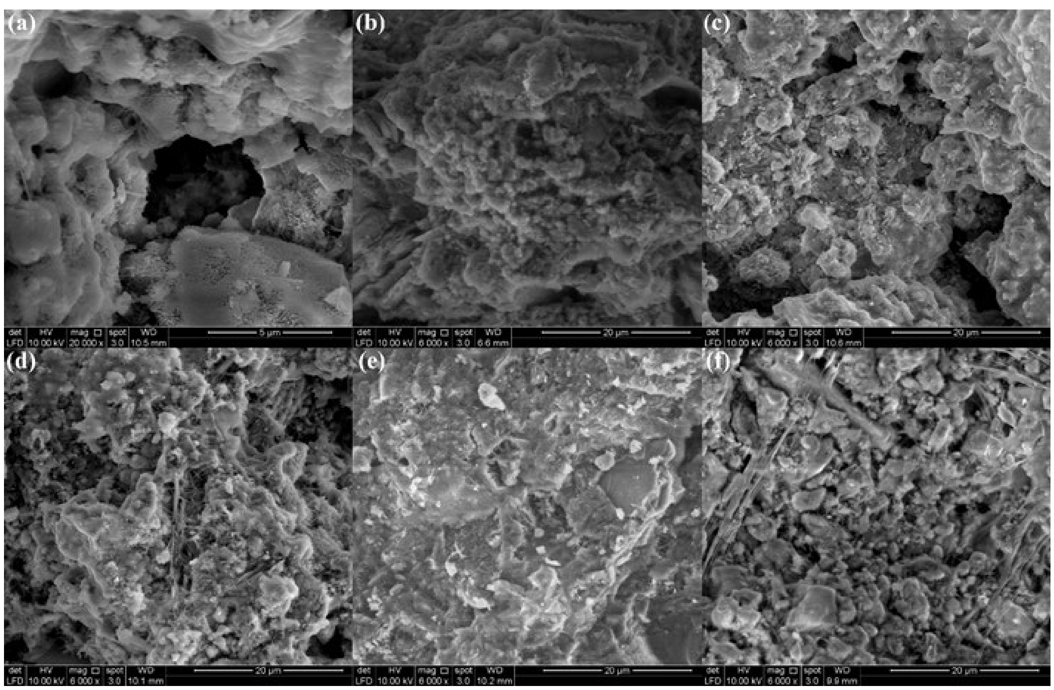

FIGURE 3. FESEM images of surface morphology (a) control brick, (b) GO composite brick 1, (c) PLCNF composite brick, (d) SCBCNF composite brick, (e) PL/GO composite brick and (f) SCB/GO composite brick.

Meanwhile in Figure 3(c) and (d), there is presence of CNF and cellulose nanocrystal (CNC) in the bricks. CNF possesses a hydrophilic structure, which is one of its characteristics is that CNF can chemically trap calcium ions in the Portland cement used [5,6]. According to Goncalves [7], the pore water is presumably trapped on the surface of CNF due to nature of carboxyl [COOH]-groups, increasing the water reservoir surrounding the CNF. Figure 3(e) and (f) shows the surface morphology of PL/GO and SCB/GO composite brick respectively and displays bricks with smoother surface and less porosity. The morphological surface of the bricks influenced by the hydroxyl groups in the CNF has reacted to the presence of GO. In addition, GO is opted for the nucleation site and densify the micropore due to its nanosized structure [8].

Radon Concentration and Porosity Analysis

In Figure 4, the highest radon concentration was recorded in control brick with reading of 67.1 Bq/m^3. Meanwhile, the lowest radon concentration is 31.2 Bq/m^3 which is from GO composite brick 1. Radon gas concentration of the brick influenced by the porosity size of the hardened cement [9]. The porosity results for the highest and lowest radon concentration of each type of bricks was illustrated in Figure 5.

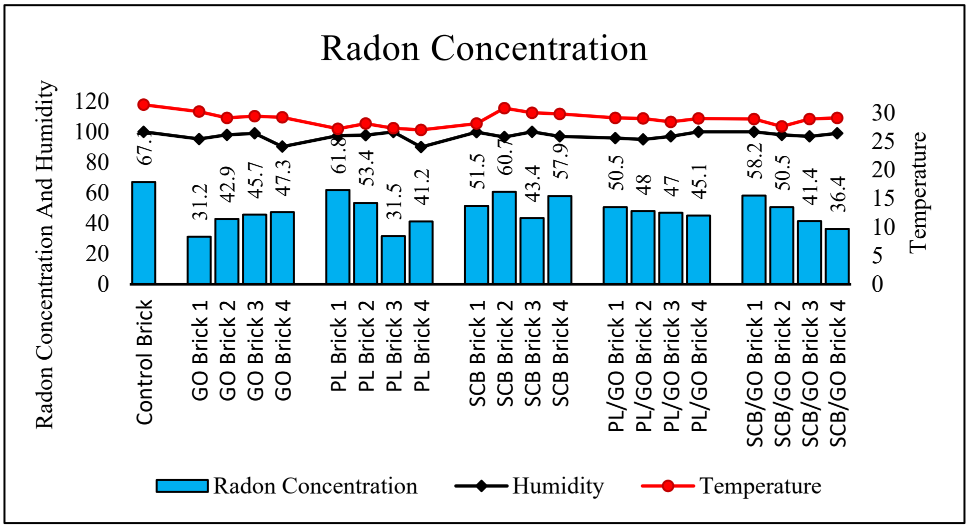

FIGURE 4. Radon concentration for control brick and composite bricks.

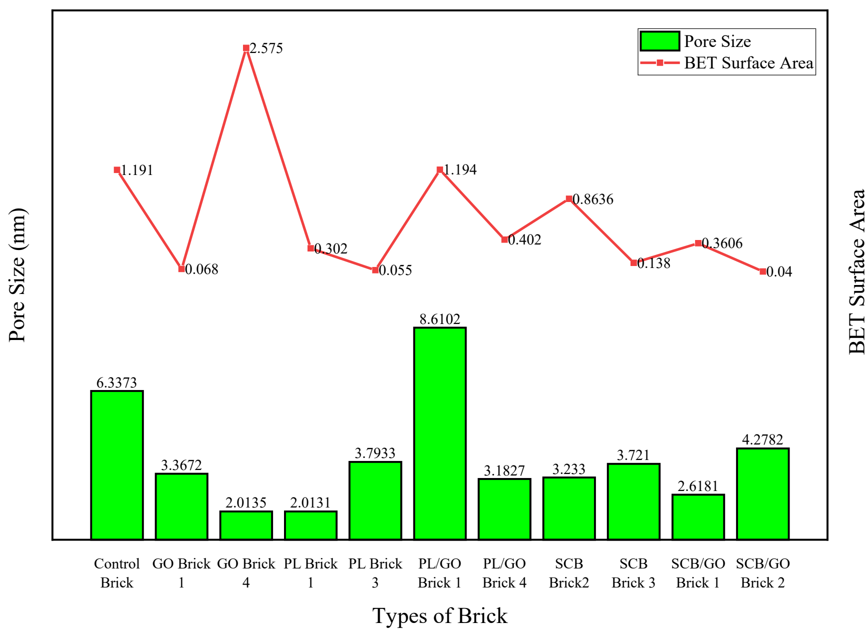

FIGURE 5. BET analysis for porosity size and surface area of the bricks.

On the other hand, GO composite brick 4 was discovered to produce the highest amount of radon with 47.3 Bq/m^3. From the results obtained, radon concentration can be reduced up to 49% by adding approximately 0.2 wt% GO to the cement mixture. This can be proved from the porosity results in Figure 5. GO composite brick 1 has the smallest pores size compared to GO composite brick 4 with porosity size 0.068 nm and 2.575 nm, respectively. GO absorb water molecules and ions generated in the hydration process and acts as a nucleation site and produces more conducive substrate for growth in hydration products and reduces the porosity of the hardened cement paste [4,10]. An excessive amount of GO results in a longer hydration period, and it will increase the formation of the water reservoir and pores [11].

Moreover, for organic composite bricks, PLCNF composite brick 3 was found to have the lowest radon concentration, which was 31.5 Bq/m^3. Meanwhile, SCBCNF composite brick 3 has the lowest radon value recorded, which is 43.4 Bq/m^3. When the volume of CNF is less than or greater than 2.1 wt% CNF, the radon concentration tends to increase. This is due to the wider porosity size and high agglomeration in the cement matrix [12]. Both bricks exhibit the smallest pore size with the reading PLCNF composite brick 3 is 0.06 nm for and SCBCNF composite brick 3 is 0.14 nm as shown in Figure 5. Inadequate homogeneity of CNF will cause higher water contents within the aggregates in the fabrication process [6]. Other than agglomeration, the increase in CNF amount causes the extreme degree of hydration which will affect the pore refinement and sizes [6,7].

For carbon organic composite bricks, the radon level in PL/GO composite brick 4 is the lowest at 45.1 Bq/m^3, and the highest is 50.5 Bq/m^3 in PL/GO composite brick 1. The lowest radon concentration was observed in SCB/GO composite brick 4 at 36.4 Bq/m^3, while the highest was found in SCB/GO composite brick 1 at 58.2 Bq/m^3. Both types of CNF/GO composite brick emanate the lowest radon concentration. It was discovered that the interaction between sufficient CNF and GO will prevent their folding with one another [13]. It might be because of GO produced has caused realignment of in-plane orientation, which makes CNF bundles more uniformly straight and reduce the agglomeration [14]. According to the results, radon was reduced approximately 33% by adding 2.8 wt% of CNF and 0.8 wt% of GO. SCB/GO composite brick 4 has an average pore size of 0.04 nm, the smallest pore size compared to other bricks. As shown in Figure 4, the findings are comparable with the results of radon in between SCB/GO composite bricks where it shows the lowest radon concentration. Compared to SCB/GO composite brick 1, it obtained bigger pores size with 0.36 nm. GO can minimize the stacking of the CNF and keep the material well separated in between GO and CNF [14]. According to a previous study done by Mianehrow [15], the reduction in brick porosity was caused by the increase in GO quantity and the presence of CNF.

CONCLUSION

From this work, it can be concluded that the composite bricks added with GO, PLCNF, SCBCNF, PL/GO and SCB/GO are able to reduce radon concentration. The optimized level for all types of composite bricks can be observed from the results. The optimized amount for GO in the cement mixture is 0.2 wt% GO, where Radon concentration has been reduced up to 49%. Meanwhile, the optimized amount of CNF in the cement is 2.1 wt% CNF. A greater or lower amount than 2.09 wt% CNF caused higher radon concentration due to wider porosity size and high agglomeration in the cement matrix. CNF/GO composite bricks with the highest weight percentage of CNF and GO can reduce the porosity of the bricks due to its combination that leads to less folding and stacking with each other. Organic and carbon-based bricks as well as the combination of both materials are feasible to reduce the radon levels from the building bricks.

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