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# Preliminary analysis of gamma and neutron irradiation on sand and activated carbon by using SEM, XRD and SAXS

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**Abstract.** Gamma and neutron irradiation effect on sand and activated carbon were studied in this paper intensively and, were found from various studies to have strong correlation between neutron fluence or gamma energy on physical and mechanical damage of materials. Sand and activated carbon were irradiated with neutron at Reactor TRIGA PUSPATI (RTP) and gamma at Sinagamma Malaysia Nuclear Agency (Nuklear Malaysia). Morphological analysis was carried out on the irradiated elements using the SEM, XRD and SAXS which showed changes on the microstructure of irradiated samples. Irradiation of activated carbon showed increase in the numbers of pores while change of textural profile of the surface take place at gamma irradiated sand. XRD pattern graph did not indicate any changes, however the specific surface area for both irradiation materials decreased.

## 1. Introduction

The National Security Council (NSC) of Malaysia secretary Datuk Mohamed Thajudeen confirmed that the massive floods which hit Kelantan in 2014 was among the worst in history. In these times of tragedy, people in affected areas usually suffer from shortages of clean water and electricity. Due to the limitations of the transportation network during emergency, only limited amount of clean water supplies could be sent to evacuation centres. As a result, some people who were familiar with water filtration often resorted to building simple home-made water filtration system using empty bottles and filter elements (cotton/coffee filter, sands and gravels)[1]. The rate of producing clean water from the flood water using this method is rather slow as several cycle filtering are required before the water can be used. This can be improved by using better filter materials. One of the ways of bettering the existing filter materials is to modify the properties of the materials. Studies had been done previously where silica (closest form of compound to sands or gravels that are commonly used as filters) was irradiated with neutron and gamma [1][2]. The effect of radiation to the adsorption and mechanical properties of activated carbon was also



studied [3]. Several authors observed that irradiation could introduce defects which resulted in the formation of micropores and different levels of irradiation level would result in different concentration of defects [4][5]. It is established that higher amount of irradiation results in higher concentration of defects in the irradiated materials[2].

## 2. Methodology

A Scanning Electron Microscope (SEM) was used to examine the surface structure (micropore) of non-irradiated and irradiated sand and activated carbon. The samples were coated with platinum to enhance their image visibility. X-Ray Diffraction (XRD) analyses were carried out using 6000 Shidmadzu, Japan at Universiti Tenaga Nasional (UNITEN). XRD patterns were measured with the following conditions: Cu K-alpha, ( $\lambda = 0.15418$  nm), tube voltage of 30 kV, current of 20 mA, scanning speed of  $4^\circ/\text{min}$  and  $2\theta$  of  $20^\circ$  to  $75^\circ$ . The results were analysed by using X'Pert Highscore Plus software. The surface area of samples was examined through Small Angle Scattering of X-Ray (SAXS) to characterize the porosity of the samples. The SAXS uses X-ray as a beam to penetrate the nanostructured material. The process took three frames where each of frame lasted for five minutes. The scattering pattern was further analysed in results and discussion. Table 1 and Table 2 describe the sample properties and the testing information of the SAXS procedure.

**Table 1.** Sample properties.

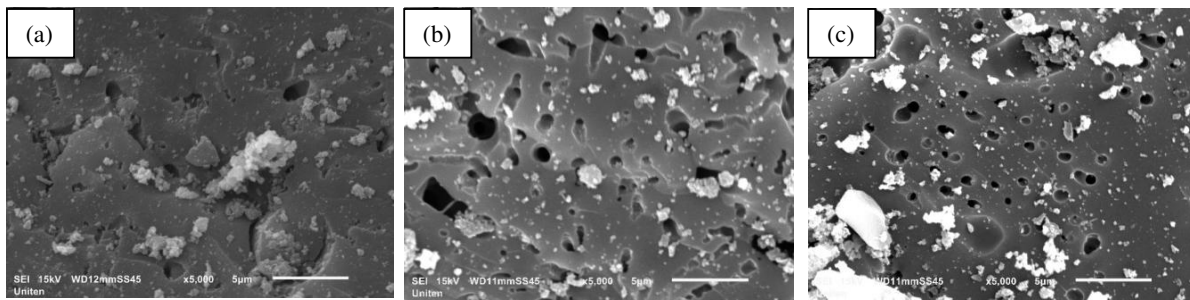
No	Sample	Form	Solid Density	Apparent Density
1	Non-irradiated Activated Carbon	Powder	2.26 g/cm <sup>3</sup>	0.58 g/cm <sup>3</sup>
2	Activated Carbon (Gamma)	Powder	2.26 g/cm <sup>3</sup>	0.58 g/cm <sup>3</sup>
3	Activated Carbon (Neutron)	Powder	2.26 g/cm <sup>3</sup>	0.58 g/cm <sup>3</sup>
4	Non-irradiated Sand	Powder	2.65 g/cm <sup>3</sup>	1.09 g/cm <sup>3</sup>
5	Sand (Gamma)	Powder	2.65 g/cm <sup>3</sup>	1.09 g/cm <sup>3</sup>
6	Sand (Neutron)	Powder	2.65 g/cm <sup>3</sup>	1.09 g/cm <sup>3</sup>

**Table 2.** Testing information.

Testing Information	Details
Sample	1-6
X-rays	Cu-K $\alpha$ , $\lambda = 0.1542$ nm
Power	50W (50 kV, 1 mA)
Sample size	0.5 mm (thickness)
Detector	2D Eiger R Series (Dectris), 75 x 75 $\mu\text{m}^2$
Calibration	Ag-behenate ( $d = 58.38 \text{\AA}$ )
Measured q-range	$0.035 \text{ nm}^{-1} < q < 5 \text{ nm}^{-1}$ (SAXS) $0.035 \text{ nm}^{-1} < q < 23.5 \text{ nm}^{-1}$ (WAXS)
Temperature	20°C
Exposure time	180 s

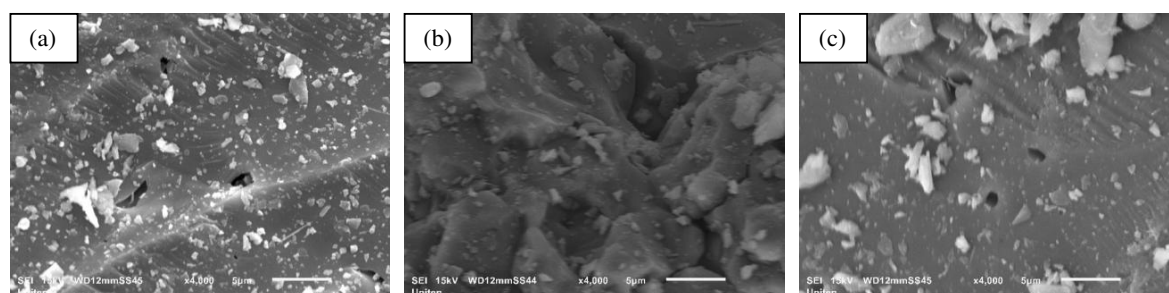
### 3. Results and discussion

The surface morphology of the non-irradiated and irradiated by gamma and neutron was evaluated by scanning electron microscope (SEM) to confirm the effect of gamma and neutron treatment on the sand and activated carbon. Figure 1 shows SEM images of the non-irradiated activated carbon, gamma activated carbon, and neutron activated carbon in 5000 $\times$  magnification. These images show the changes on their surface structure after irradiation. It can be observed that there were many pores formed on them. This was due to the ability of gamma-rays and neutrons to cause atomic displacement [13]. They interacted with carbon nucleus mainly through Compton scattering and absorption. The recoiled electrons with sufficiently high energy from the Compton scattering would collide with neighbouring carbon nuclei, shifting them from their lattice position. Then, the knocked-on atoms would continue to collide with their neighbouring atoms, resulted in transferring of energy. If the energy exceeded the threshold of further interactions and displacements, it would result in cascading damage [33]. The average size of pores was taken using dimension tools provided by the SEM where pores were chosen at random. The neutron-radiated activated carbon was average pore diameter of 0.7881  $\mu\text{m}$ . Meanwhile, the non-irradiated activated carbon and gamma-radiated activated carbon showed no significant difference in average pore diameters which were 1.429  $\mu\text{m}$  and 1.603  $\mu\text{m}$  respectively.



**Figure 1.** SEM image at (5000 $\times$ ) magnification of: (a) non-irradiated activated carbon, (b) irradiated activated carbon with 50kGy gamma dose, (c) irradiated activated carbon with  $2.000 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$  neutron flux.

Figure 2 shows the SEM images of non-irradiated and irradiated sand at 4000 $\times$  magnification. Generally, sand (mainly build-up from quartz) has a particle shape with higher sphericity index and lower internal porosity [3][4]. The SEM image for gamma-radiated sand in Figure 4.2(b) shows the change in textural profile of its surface resulting valley and hill areas as compared to the non-irradiated sand in Figure 4.2(a) and the neutron-radiated sand in Figure 4.2(c). Pores were only visible on the surface of the non-irradiated sand and the neutron-radiated sand where their average sizes were 1.334  $\mu\text{m}$  and 1.043  $\mu\text{m}$  respectively. The change in topography of the surface and the small average size of pores prove that irradiation of neutron and gamma-ray can induce micropores or defects in the materials [4][5].

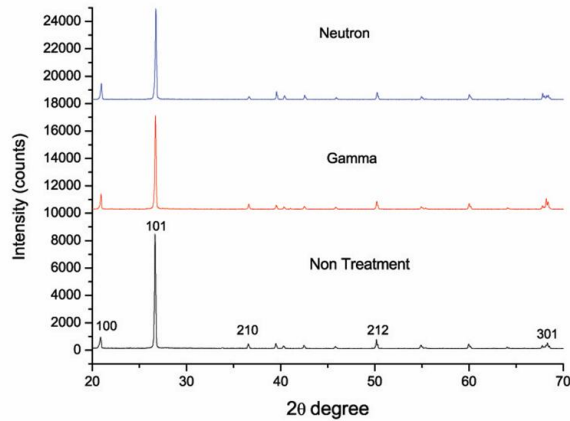
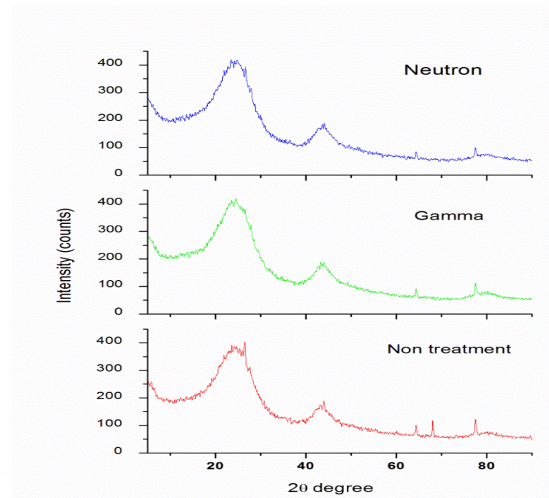


**Figure 2.** SEM image at (4000 $\times$ ) magnification of: (a) non-irradiated sand, (b) irradiated sand with 50 kGy gamma dose, (c) irradiated sand with  $2.000 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$  neutron flux.

Based on the discussion above, the neutron-radiated activated carbon has the smallest average pore size of 0.7881  $\mu\text{m}$ . This might contribute to the highest capability of filtration as the small pores could trap foreign and contaminated substances in the water. The XRD analysis was performed to investigate the structure and crystallinity of the non-irradiated and irradiated sand and activated carbon which are presented in Figure 3 and Figure 4 respectively. It is known that if the radiation has high enough energy, it can cause build-up of defects on the materials. This may collapse the crystalline lattices to the extent of inducing crystalline-amorphous transition due to the existing of vacancies-interstitials pair [3].

Based on the XRD profiles, the non-irradiated sand shows the highest peak which indicates that it is highly crystallized with 011 hkl at  $2\theta$  of  $26.6586^\circ$ . This suggests that it has hexagonal structure [4]. Using Debye-Scherrer equation on high intensity peak, the average crystallite size of particles was calculated to be 62.39 nm. Apparently, the XRD pattern for irradiated sand did not show substantial changes, following the same pattern as the non-irradiated sand. However, there were nominal changes in the average crystallite size of gamma-radiated sand and neutron-radiated sand which were 68.46 nm and 64.50 nm. Compared to non-irradiated sand, both of the irradiated sand samples had slightly small crystallite size which might improve their filtration effectiveness [5][6].

Figure 4 illustrates the XRD spectra for the activated carbon samples. The diffractogram pattern of these samples exhibits two distinctive broad reflections in the  $2\theta$  angles approximately at  $25^\circ$  and  $44^\circ$ . The peak broadening is an indication of an amorphous structural aspect. The broad reflection in the  $2\theta$  at  $25^\circ$  and  $44^\circ$  are associated with (002) and (001) diffraction line respectively. The similar pattern shown concludes that nothing had changed in the amorphous structure of activated carbon after being irradiated by gamma and neutron. This happened because radiation does not carry sufficient energy to cause the change [3].

**Figure 3..** XRD of Sand.**Figure 4.** XRD of Activated Carbon.

However, a previous study showed that ionising radiation can cause loss of crystallinity or loss of weight in graphite. This stems from the breakage of the chemical bonds which results in the creation of defects and in the loss of regular structure. In the case of persistent or higher concentration of radiation, a fraction of the defects condenses and may form extended defects by arranging carbon atoms in the basal layers [7].

A sample of non-irradiated sand was evaluated with SAXS to identify the nanosized particle. When X-rays penetrate a nanostructured material, they are scattered on the interfaces of the nanostructures at a certain scattering angle. The resulting scattering pattern is unique to the structure and this is used to characterize the nanomaterial. The data were processed with SAXStreat and SAXSquant softwares. Then, evaluation was performed with the SAXSquant and SGI (Space group indexing). Table 3 shows specific surface area value for each sample.

**Table 3.** Specific Surface area, Si value.

Sample	Si (m <sup>2</sup> /g)
Non-irradiated Activated Carbon	1437.5
Gamma Activated Carbon	863.6
Neutron Activated carbon	679.9
Non-irradiated Sand	122.6
Gamma Sand	104.6
Neutron Sand	81.89

Based on Table 3, the specific surface area of the sample decreased after irradiation. As the area decreased, the particle size became small. Specific surface area can also decrease if the sample has less pores [8]. These results seem to agree with a previous study which mentioned that the gamma-irradiated activated carbon had reduced specific surface area and micropores volume [9]. Gamma-rays interactions with bulk materials are different from that of charged ions and neutrons. It is known that the probability

of photoelectric interaction and pair production is insignificant for the case of highly-energetic gamma-rays and absorbing material of low atomic number such as graphite. Therefore, Compton scattering is believed to be the major interaction mechanism of high-energy photons with the carbon atoms in graphite lattice [7]. Another reason for the decreasing specific surface area of sand is due to the energy transferred to the quartz, resulting in the knocking and breaking of some of the porous wall. This blocks the porosity formation, disturbing the surface area of micropores [10]. Table 4 shows the summary of radiation effects of sand and activated carbon

**Table 4.** Summary of radiation effects.

Analysis Sample	Morphology	Structure	Surface Area (m2/g)		
Sand	Neutron	<ul style="list-style-type: none"><li>•small number of pore does visible in SEM image</li><li>•have small average pore diameter = 1.043µm compared to non-irradiated (1.334 µm )</li></ul>	<ul style="list-style-type: none"><li>•The XRD shows same pattern throughout all types of samples.</li><li>•Nominal change in average crystallite size irradiated sand with gamma is 68.46nm</li></ul>	<ul style="list-style-type: none"><li>•81.89 m2/g</li></ul>	<ul style="list-style-type: none"><li>•Specific surface area were decreasing after irradiate with gamma and neutron from non-irradiated is 122.6m2/g</li></ul>
	Gamma	<ul style="list-style-type: none"><li>•change of textural profile of the surface compared to non-irradiated and neutron sand</li><li>•pore does not visible in SEM image</li></ul>	<ul style="list-style-type: none"><li>•The abundant element is SiO2 (quartz) indicates the hexagonal structure</li><li>•irradiated sand with neutron is 64.50nm (non-irradiated =62.39nm</li></ul>	<ul style="list-style-type: none"><li>•104.6 m2/g</li></ul>	
Activated Carbon	Neutron	<ul style="list-style-type: none"><li>•Increasing number of pores</li><li>•have small average pore diameter = 0.7881µm compared to non-irradiated and gamma sample</li></ul>	<ul style="list-style-type: none"><li>•The diffratogram pattern of these samples exhibits 2 distinctive broad reflections in the 2θ angles located approximately at 25° (002) and 44° (001).</li></ul>	<ul style="list-style-type: none"><li>•679.9 m2/g</li></ul>	<ul style="list-style-type: none"><li>•Specific surface area were decreasing after irradiate with gamma and neutron from non-irradiated is 1437.5 m2/g</li></ul>
	Gamma	<ul style="list-style-type: none"><li>•Increasing number of pores</li><li>•average pore diameter (1.429 µm )does not have significant change compared to non-irradiated activated carbon (1.603 µm )</li></ul>	<ul style="list-style-type: none"><li>•The peak broadening is an indicative of an amorphous structural aspect.</li></ul>	<ul style="list-style-type: none"><li>•863.6 m2/g</li></ul>	

#### 4. Conclusions

It was hypothesised, based on previous studies, that the irradiation of sand and activated carbon might defect the structure of materials with sufficient energy and time. In this research, the effect of gamma-rays and neutrons to sand and activated carbon were analysed using SEM, XRD and SAXS. Different radiation treatment produced different effect to the structure of the materials[2]. We observed more irradiation energy is required to obtain further understanding related to structural changes in the samples.

## 5. References

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