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Comparison on characteristic of Mesoparticle Graphene Sand Composite (MGSC) using different types of sugar to remove methylene blue

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Abstract. This paper presents the green method to synthesis two types of adsorbent called mesoparticle graphene sand composite (MGSC) by using table sugar (MGSC_{ts}) and arenga palm sugar (MGSC_{aps}) as different carbon sources to remove methylene blue acted as a dye model. Immobilisations of these materials on sand were introduced by using pyrolysis method without binder usage. Sand was treated by removing deleterious materials before sieved. The solutions of sugar were prepared and heated to 95 °C. The sand and sugar solutions were mixed and constantly stirred before putting them in furnace with nitrogen environment to produce MGSC_{ts} and MGSC_{aps}. The composites were activated by using concentrated sulphuric acid to open the pores and maximise the capacity of absorbency. The analyses on the characteristic of both MGSC_{ts} and MGSC_{aps} were conducted through field emission scanning electron microscope (FESEM), elemental dispersive x-ray (EDX) and elemental mapping (EM). FESEM analyses exhibited that the coating process was done uniformly as there were layers of coating sheets formation on the sand particles surfaces. After conducting EDX and EM, there were major elements found in both MGSC_{ts} and MGSC_{aps} which were carbon, oxygen and silica. EM exhibited the distribution of these elements were scattered on the MGSC's surfaces. Removal of methylene blue was successfully carried out by using both MGSC_{ts} and MGSC_{aps}, with maximum removal up to 40% at the first hour of contact time.

1. Introduction

The applications of composite materials in contaminated air, water and soil are intensely pursued recently [1], [2] as the released of industrial wastewater to the clean water sources is very difficult to manage and indirectly caused imbalance ecosystem. Thus, it is necessary to find a way to treat [3] and purify waste water [2] to produce cleaner waste before releasing it to the environment. High adsorption of waste in wastewater can be achieved by using a suitable and solid sorbent. The changes in the earth's climate have negative impacts on water resources around the world [4] which lead to the increasing periods of drought, more floods, higher water temperatures, and changes in water chemistry [5] [6]. Hence there is a significant need for the development of efficient and economical wastewater treatment technologies. This paper presents on the synthesis and characterisation process of green mesoparticle graphene sand composite (MGSC_{aps} and MGSC_{ts}) to remove methylene blue from wastewater by using arenga palm sugar and table sugar respectively as carbon sources.



Waste material such as rubber tyres [7] and other adsorbent materials [8], [9], [10] are also been used in adsorbing heavy metals and dyes. The removal is efficient; however the cost is high to synthesis and produces it as a good adsorbent. Thus, the researches on the green composite with facile, lower cost of synthesisation and production are a need for environmental remedy. Furthermore, the additional interesting characteristic of the green and facile adsorbent is the reusability and regeneration of the composite in the next cycle of adsorption process. Recently, there is one composite material that getting paid attention, namely graphene. Hence, the fundamental knowledge of the characteristic, absorbance process and the possible failure of the system are essential before any remediation work is carried out. In this article, the main discussion and review is made on the characteristic and synthesis of graphene, and the potential of MGSC as absorbent to remove methylene blue in wastewater. The objectives of the research are to synthesis MGSC using green technologies and characterise it before being tested as adsorbent. Arenga palm sugar and table sugar were chosen to be acted as the main source of carbon since there were high in sucrose so far. Thus the overall performance of MGSC_{aps} and MGSC_{ts} in removing pollutants needs to be investigated.

2. Methodology

2.1. Materials

The materials used to synthesis MGSC_{aps} and MGSC_{ts} were arenga palm sugar and table sugar as carbon sources, and sand which passed 2 nm sieve. Arenga palm sugar was obtained from arenga pinnata tree which abundantly available regions around the world especially in the tropical such as Malaysia, Indonesia, Thailand, Philippines and Vietnam. In this research, arenga palm sugar was supplied from Pahang, Malaysia whereas table sugar was supplied by Prai Sugar Sdn Bhd. Sand was collected from river located in Pahang, Malaysia.

2.2. Chemicals

Methylene blue and concentrated sulphuric acid were supplied from Permula Chemicals Sdn. Bhd. Distilled water as solvent was used throughout this experimental process.

2.3. Instrumentation for MGSC_{aps} Characterisation

MGSC_{aps} and MGSC_{ts} were subjected to several types of testing including FESEM, EM and XRD.

2.3.1. Field Emission Scanning Electron Microscope. Field emission scanning electron microscopic (FESEM) was analysed by using FESEM JEOL JSM-6500F equipped with MiniFlex2 counter detector. It was also used fixed monochromator of diffracted beam monochromator.

2.3.2. Elemental Mapping. The growth and distribution of carbon on silica oxide surfaces was recorded by using elemental mapping equipment JEOL JSM-6500F.

2.3.3. Elemental Dispersive X-Ray. Elemental Dispersive X-Ray (EDX) was recorded to ascertain elements percentage of graphene sand composite by using 30 kV; 15 mA x-ray with fixed monochromator at 1.0000 sec scan speed.

2.4. Synthesis Process of MGSC_{aps} and MGSC_{ts}

The synthesisation of MGSC_{aps} and MGSC_{ts} followed method mentioned in the literature [11] with a slight modification on the carbon source. For MGSC_{aps}, 150 g arenga palm sugar was used as carbon source and was diluted in 150 g distilled water. The solution was then mixed with 150 g fine river sand before constantly stirred at 95 °C for 3 hours. The mixture then was put in the silica crucibles and burned in the nitrogen atmosphere furnace. The temperature of the furnace was set from the room temperature to 100 °C in 30 minutes to burn deleterious materials. The temperature was changed to 200 °C in another 30 minutes and was held for 1 hour to melt arenga palm sugar to confirm uniform

coating on the sand surfaces. The temperature of the furnace then was ramped to 750 °C in 1 hour and held it for another 3 hours to ensure graphenisation process was completed. MGSC_{aps} then allowed cooling to the room temperature gradually. After cooled, MGSC_{aps} activated by using concentrated sulphuric acid for 30 minutes without disturbance to open pore sites which acted as adsorbent sites. MGSC_{aps} is now ready for adsorption process before filtered as shown in Figure 1. The preparation of MGSC_{ts}, was followed the same step as MGSC_{aps}'s preparation. However the source of carbon was changed from arenga palm sugar to table sugar.

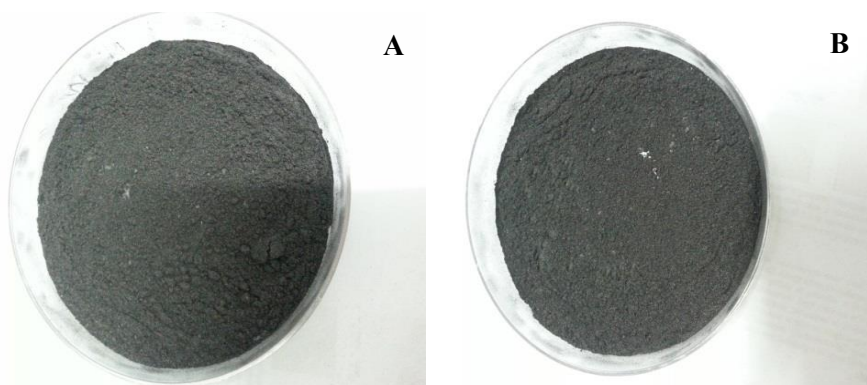


Figure 1. MGSC_{ts} (A) and MGSC_{aps} (B) were successfully synthesised

3. Results and Discussion

3.1. MGSC_{aps} and MGSC_{ts} Characterisation

3.1.1. Field Emission Scanning Electron Microscope. The morphological of MGSC_{aps} and MGSC_{ts} were carried out through FESEM analyses. It was evident that the sand was coated uniformly which confirms the temperature setting for initial burning process was 200 °C successfully melted arenga palm sugar to form a layer of carbon coating. This is in line with the previous researchers [12] which stated that the melting point of sucrose is approximately 186 °C. The graphenic morphology also was detected at the coating sheet's edges where thin wrinkles were formed as shown in Figure 2. The wrinkled edges of sheets that confirms the production of graphene, even after the formation of the composites, the structure of the composite remained the same where the wrinkles were still present [13].

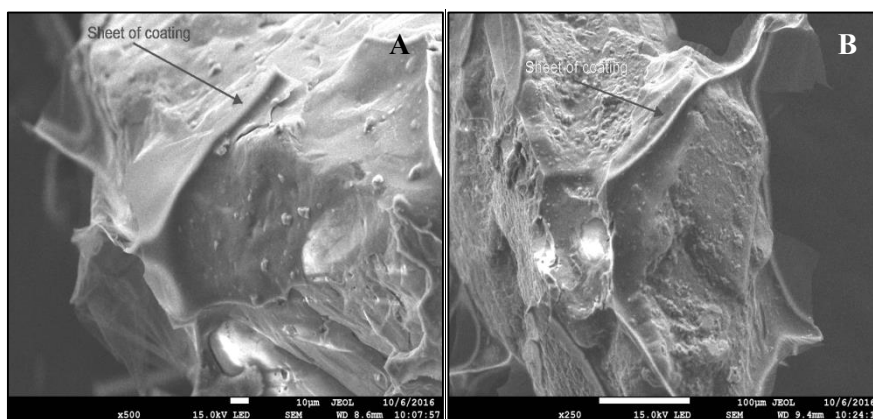


Figure 2. Sheet of coating was formed after arenga palm sugar (A) and table sugar (B) melted

3.1.2. Elemental Mapping. Major elements present were carbon, silicon and oxygen. The presence of large carbon feature was originated from the carbonaceous material which was formed from melted arenga palm sugar and table sugar on sand surface. Previous researchers [14] stated that the elements detected show that the sand was covered by carbon. The other elements; silicon and oxygen were also features in the major elements due to the usage of sand (SiO_2) hence validating the name of this composite in the research, mesoparticle graphene sand composite (MGSC_{aps} and MGSC_{ts}).

3.1.3. Elemental Dispersive X-Ray. An EDX analysis was conducted to measure the percentages of each chemical element in MGSC_{aps} and MGSC_{ts} . Figure 3 presents the result of MGSC_{aps} and MGSC_{ts} upon elemental dispersive x-ray test. From the results, there were major chemical elements' peaks in line with EM results which were carbon, silicon, oxygen. Other chemical elements' peaks were ferum and aluminium found on sand surfaces.

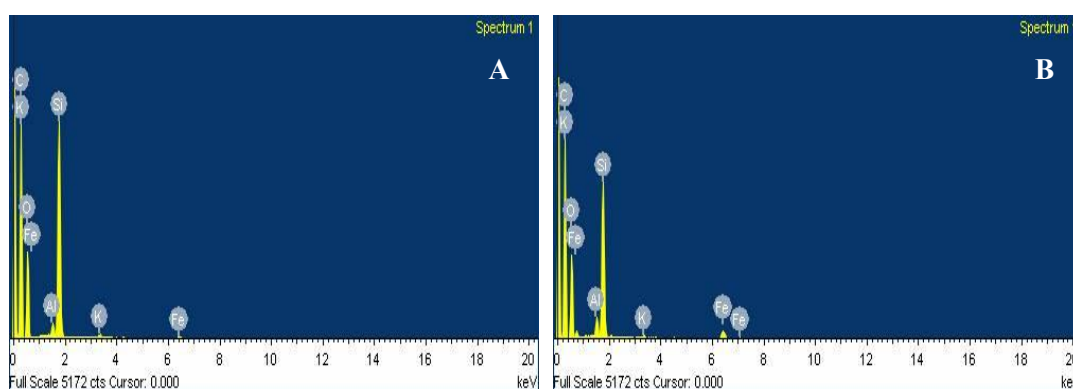


Figure 3. The chemical elements' peaks of MGSC_{aps} (A) and MGSC_{ts} (B)

4. Conclusion

The green adsorbents by using arenga palm sugar and table sugar were successfully synthesised. The chemical elements in the both adsorbent was confirmed the relevance of the composite name which is mesoparticle graphene sand composite (MGSC). Based on the characteristics of the composite, it can be concluded that it has significant character to be acted as a good adsorbent in removing methylene blue.

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