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Zinc oxide functionalized biochar from rambutan wood as a novel material to utilize an agricultural waste in Thailand

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Abstract

Zinc oxide-functionalized biochar (biochar-ZnO) from rambutan wood, an agricultural waste, was prepared to increase a value of biochar from rambutan, an important economic fruit crop in Thailand, for the first time. Biochar-ZnO was synthesized by a sol-gel process. X-ray diffraction (XRD) spectra suggest that the biochar consists of amorphous carbon mainly and confirm the success of zinc oxide functionalization on the biochar surface. The crystal structure of zinc oxide is hexagonal Wurtzite. The diameter of zinc oxide particles is ~20 nm. Thermogravimetric (TGA) thermograms indicate the success of biochar-ZnO preparation. A weight percentage of zinc oxide in the composite is ~20. Their Fourier transform infrared (FTIR) spectra confirm, again, the success of ZnO functionalization on the biochar surface and indicate both carboxylic and hydroxyl groups on the biochar and biochar-ZnO surface. The iodine numbers of the biochar and biochar-ZnO are 570 and 555 $mg\cdot g^{-1}$, respectively. These numbers suggest that the biochar without an activation has a surface area that is comparable to that of activated carbon. Also, a decrease in the iodine numbers after the sol-gel process suggest a coverage of zinc oxide particles on the biochar surface. Lastly, biochar-ZnO is a promising material to be used for further applications such as a filler in rubber industry, an important industry in Thailand.

1. Introduction

Rambutan, a native fruit from Southeast Asia, is a Thai important economic fruit crop from Southern Thailand and a coastal area of Eastern Thailand (Chanthaburi, Rayong, and Trat) [1]. According to Thailand Department of Internal Trade, Ministry of Commerce, a production of rambutan from Thailand in 2024 is 219,000 metric tons. This leads to a significant amount of rambutan residues, including rambutan wood.

Normally, rambutan gardeners will simply burn rambutan wood to obtain charcoal when rambutan trees cannot give fruits anymore as a way to manage waste. Charcoal from a biological source or 'biochar' is an aromatic carbon-rich solids, which is produced by a low-cost and relatively low temperature pyrolysis from a renewable resource or agricultural wastes such as rice straw, wheat straw, and corn straw [2]. Biochar has several interesting properties such as high specific surface area and porosity, high pore volume, good water-holding capability, good ion exchange capability, and high heating value [2]. Therefore, biochar can be used in various applications such as catalysts for energy production, biofuel production, waste management, control of air pollutants, and energy storage. It can also be used in remediation of pollutants, soil amendment, carbon sequestration, and wastewater treatment [3].

Although biochar can be used in a number of applications, in practice, it is normally used as a fuel for cooking and as an improving soil matter. Unfortunately, a price of biochar from rambutan wood is not high. Therefore, we have an idea to increase a value of inexpensive biochar by treating it chemically to obtain more valuable materials.

Zinc oxide is a metal oxide that is usually used with carbon black in rubber industry for sulfur vulcanization of rubber [4], as a pigment in rubber [5], for enhancing rubber durability [6], for imparting germicidal properties in rubber [7], for catalysis [8], for UV protection in sunscreen, and for environmental remediation [9].

Due to the utility of zinc oxide and biochar, there are attempts to prepare composites between them such as ball-milled synthesized biochar-zinc oxide nanocomposite using Zea mays L. [10] and zinc oxide betaine-modified biochar nanocomposites [11]. The composites between zinc oxide and biochar were also prepared to be used as effective catalysts for electrochemical CO_2 reduction [12] and controlled release of fertilizer [13].

Although the composites between biochar and zinc oxide were prepared, biochar from different sources have different characteristics [2]. To the best of our knowledge, there is no published research work on biochar from rambutan wood. Therefore, this present work aims to increase a value of biochar from rambutan wood by a chemical functionalization with a zinc compound to obtain zinc oxide

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functionalized biochar from rambutan wood (biochar-ZnO) for the first time.

2. Materials and methods

2.1 Materials

Ascorbic acid (Northeast Pharmaceutical Group, Liaoning Province, P.R.C.), biochar from rambutan wood (Nikompattana market, Nikompattana district, Rayong province, Thailand), iodine (resublimed AR, Loba Chemie, India), potassium dihydrogen orthophosphate anhydrous (98%, Loba Chimie, India), potassium iodide (AR, 99%, Loba Chimie, India), sodium hydroxide (99%, AR grade, AGC Chemicals (Thailand), Thailand), sulfuric acid (RCI Labscan, Thailand), and zinc sulfate heptahydrate (99%, AR grade, Loba Chimie, India) were used as received. Purified water from Faculty of Science, Energy and Environment, King Mongkut's University of Technology North Bangkok (Rayong Campus) was used throughout the experiments.

2.2 Preparation and characterization of biochar and zinc oxide functionalized biochar (biochar-ZnO)

The biochar was prepared from rambutan wood by pyrolyzing the wood at ~700°C. and ground in the mortar to obtain small biochar particles. Both 0.1 M NaOH (100 mL) and 0.5 M zinc sulfate heptahydrate (100 mL) solutions were prepared separately. Ground biochar (1 g) was poured into the zinc sulfate solution. The mixture was stirred. Then, the sodium hydroxide solution was slowly dropped into the mixture. The mixture was continuously stirred for 2 h and filtrated. Obtained particles were washed by purified water three times and warmed in an oven at 80°C overnight. The obtained particles were characterized by TGA (Q50, TA Instruments, USA) under air with a heating rate of 10°C·min^{-1} from ambient temperature (25°C) to 800°C to examine weight percentages of ZnO in the composite and carbon content in the samples. Then, the biochar and biochar-ZnO were characterized by XRD (D8 Advance, Bruker, USA) with a Cu-Kα radiation in a range of 10° to 80° for 15 min to examine a crystallinity of carbon and ZnO in the samples and ZnO phase the composite. Functional groups on the biochar and composite surface were studied by ATR-FTIR (Attenuated total reflectance-Fourier transform infrared spectroscopy, Frontier, PerkinElmer, USA) within a range of 4000 cm^{-1} to 400 cm^{-1} and a resolution of 4 cm^{-1}. Surface area of the biochar and biochar-ZnO was evaluated by using the standard test method for determination of iodine number of activated carbon (ASTM 4607-14).

3. Results and discussion

XRD patterns of the biochar and biochar-ZnO are shown in Figure 1.

A XRD pattern of the biochar shows a broad peak at 23° It is defined as a crystal plane index C(0 0 2) that is related to a parallel orientation of an aromatic carbon structure [14]. A broad peak at 43° (C(100)) is due to carbons of graphite layers [15]. An interlayer spacing was ~0.395 nm. A character of this broad peak indicates that the biochar comprises amorphous carbon mainly in a form of graphite as a result of cellulose decomposition. A spectrum of biochar-ZnO shows diffraction peaks at 32°, 34°, 36°, 48°, 57°, 63°, 68°, and 69° that correspond crystal planes of (100), (002), (101), (102), (110), (103), (200), (112), (201), and (202) crystal planes of ZnO hexagonal Wurtzite structure (JCPDS card 36–1451). An intensity of the peaks from ZnO is higher than that of the peaks from the biochar. It means that crystallinity of carbon in the biochar is low and, thus, carbon structures in the biochar and the composite are mainly amorphous. From the Scherrer equation, an average size of ZnO particles in the composite is ~20 nm. Then, both the biochar and biochar-ZnO were subjected to TGA to analyze the compositions in the biochar and the composite. Their TGA thermograms are shown in Figure 2.

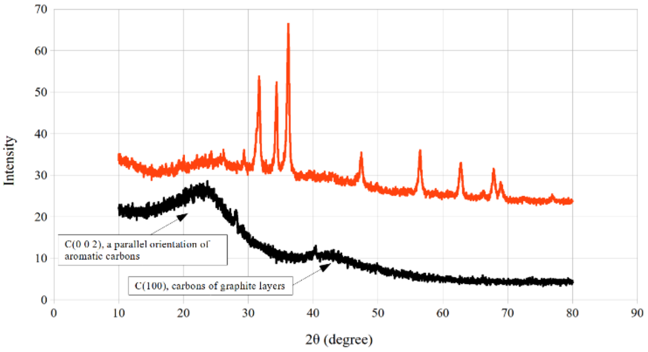

Figure 1. XRD spectra of the biochar (black) and biochar-ZnO (red).

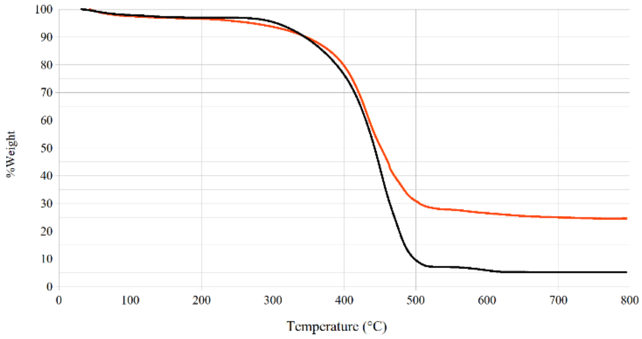

Figure 2. TGA thermograms of the biochar (black) and biochar-ZnO (red).

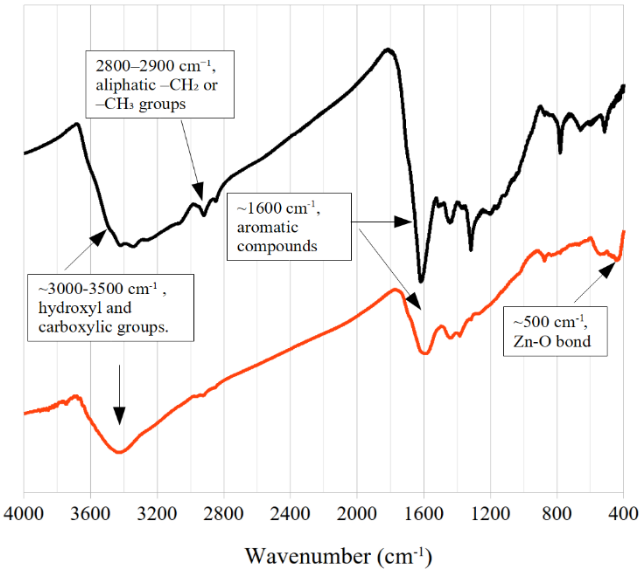

Figure 3. ATR-FTIR spectra of the biochar (black) and biochar-ZnO (red).

Figure 2 shows a < 5% weight reduction at 100°C for both the biochar and biochar-ZnO. This suggests that moisture slightly contaminated the samples. Both samples were combusted under air from 300°C to 500°C as we observe the weight reduction during this period. After 500°C, the biochar and the composite ashes weighted ~5% and ~25% of the total weight, respectively. It suggests that a weight of ZnO in the composite is ~20% and confirms the success of ZnO functionalization on the biochar surface. Both samples were subjected to FTIR for identifying the functional groups on their surface. The results are shown in Figure 3.

The FTIR spectrum of the biochar shows the characteristic band at ~1600 cm^{-1} corresponding to C=C stretching in aromatic compounds [16]. The peak at ~2800 cm^{-1} to 2900 cm^{-1} is related to C–H stretching of methylene and methyl groups in aromatic compounds. The peak at ~3000 cm^{-1} to 3500 cm^{-1} is related to O–H stretching of hydroxyl and carboxylic groups that formed during the pyrolysis process [16]. The FTIR spectrum of biochar-ZnO shows a characteristic band at ~1600 cm^{-1} corresponding to aromatic groups, whereas the peak at ~3450 cm^{-1} is related to O–H stretching in carboxylic and hydroxyl groups [16]. Therefore, the functional groups on the biochar-ZnO surface are similar to those on the biochar surface. However, the composite also shows the peak at ~500 cm^{-1} that corresponds to Zn–O bonds [11]. Also, the weak signal at ~2800 cm^{-1} to 2900 cm^{-1} that is related to C–H stretching of methylene and methyl groups in aromatic compounds disappears. It was possibly caused by the coverage of zinc oxide particles on the composite surface. This spectrum also, again, confirms the success of ZnO functionalization on the biochar surface.

Next, the iodine numbers of the biochar and biochar-ZnO are 570 $mg\cdot g^{-1}$ and 555 $mg\cdot g^{-1}$, respectively. It indicates that a surface area of the biochar without an activation is comparable to that of activated carbon, whose iodine number is in between 500 $mg\cdot g^{-1}$ and 1,200 $mg\cdot g^{-1}$. Also, a decrease in the iodine numbers after the sol-gel process was possibly caused by the coverage of zinc oxide particles on the biochar surface that is consistent with the disappearance of the weak signal at ~2800 $mg\cdot g^{-1}$ to 2900 $mg\cdot g^{-1}$ in the ATR-FTIR spectra of the composite.

4. Conclusions

TGA thermograms suggest that biochar-ZnO with ~20%w/w ZnO was successfully prepared by a sol-gel method. The XRD spectra suggest that the biochar and the composite consist of amorphous carbon. The crystal structure of ZnO in the composite is hexagonal Wurtzite and the size of ZnO particles is ~20 nm. The FTIR spectra show the presence of carboxylic and hydroxyl groups on the biochar and biochar-ZnO surface. The iodine numbers of the biochar and biochar-ZnO are 570 $mg\cdot g^{-1}$ and 555 $mg\cdot g^{-1}$, respectively. Their surface area is comparable to that of activated carbon. Also, these iodine numbers suggest the coverage of ZnO particles on the biochar surface. These results suggest that our prepared biochar-ZnO composite is potentially used in industry such as a filler in natural rubber. This leads to the increase in value of ordinary biochar from agricultural waste to a potential raw material in industry.

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Conflicts of interest

The authors declare no conflict of interest.

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