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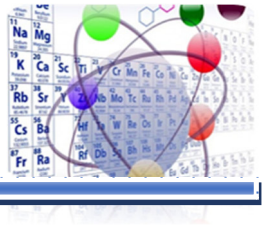

Effect of Cu and Fe Metals on the Synthesis of Activated Carbon Composites of MOFs Cu(TAC) and Fe(TAC)

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ABSTRACT

The research focuses on improving the structure and quality of activated carbon by synthesizing porous polymers or Metal-Organic Frameworks (MOFs). Characterization of AC-Cu(TAC)$_2$ and AC-Fe(TAC)$_3$ is carried out using XRD, SEM, and EDX instruments. Characterization of AC- Cu(TAC)$_2$ and AC-Fe(TAC)$_2$ revealed that the observed XRD pattern is representative of the structure of crystalline materials, with AC-Cu(TAC)$_2$ and AC-Fe(TAC)$_3$ having a crystallinity of 40% with both materials forming a triclinic crystal system. Characterization based on SEM at 200x magnification shows that the fibers are interbonded and attached to the cross-section on AC- Cu(TAC)$_2$, having a relatively large porosity so that it shows a fairly strong bond between activated carbon and Cu(TAC) MOFs. the size of the fragmented fiber particles in AC-Fe(TAC)$_3$ which is relatively not the same as the smaller porosity. EDX characterization shows that Cu and Fe metals are one of the composite materials.

Keywords: Oil Palm Empty Bunches (OPEB), Activated Carbon, Composite, AC-Cu(TAC)$_2$, AC- Fe(TAC)$_3$,

1. INTRODUCTION

Metal Organic Frameworks (MOFs) are porous coordination polymers composed of organic metals and *linkers* so that they are able to form porous crystals so that when compared to other porous materials such as activated carbon, zeolite and silica, MOFs show better user configuration flexibility through manageable pores and selectable functional groups.[1] MOFs can be widely applied in various fields including adsorption, separation, catalysis and drug delivery. This is because MOFs have high porosity (up to 7000 m^2/g).[2] MOFs used as adsorbent materials are able to provide superior surface area, uniform pore structure and have high selectivity, MIL-100(Fe) which is one of the types of MOFs using Fe metal as a constituent metal displaying a large surface area and pore volume, namely 1350 – 2300 m^2/g and 0.82 – 1.20 cm^3/g.[3]

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With the various advantages shown by MOFs, MOFs are often used as one of the composite materials. Composite is a material formed from the combination of two or more constituent materials that have different mechanical properties.[4] Various types of MOFs composites are being developed today such as MOFs – carbon-based composites, MOFs-silica-based composites, nanoparticles MOFs – metals, MOFs polymer composites – organic.[5] In MOF composites, these composites can integrate various advantageous things and compensate to cover the weaknesses of their single components. Carbon- MOF Composite is a combination of MOF and carbon-based materials. The properties of carbon-based materials, such as high mechanical strength and elasticity, as well as good chemical and thermal resistance, but have some weaknesses such as irregularities in pore size and weak adsorbate and adsorbent-adsorbent interactions. By integrating MOFs into carbon-based materials, these composites not only address the drawbacks of activated carbon, but also create a variety of new functions, including improved stability and electrical conductivity.[1]

2. EXPERIMENTAL

2.1. Chemicals, Equipment and Instrumentation

The materials used in this study include oil palm empty bunches (OPEB), TAC (*Terephtalic Acid*), $FeCl_3.6H_2O$ (99% Merck), $Cu(NO_3)_2$ (99% Merck), HF (Fluoric Acid), H_3PO_4 (Phosphate Acid), HNO_3 (Nitric Acid), Aquades. The tools used in this study include glassware, burets, statics, clamps, reflux, grinders, analytical balances, ovens, furnaces, hotplates, 200 mesh sieves, SEM, EDX, and XRD

2.2. Research Procedure

2.2.1. Empty Oil Palm Bunches Preparation

The empty oil palm bunches used were obtained from PT. Perkebunan Nusantara II Pagar Merbau. The empty bunches obtained are then washed and dried in the sun. After drying, the empty bunches of oil palm are cut into small parts and after that are ground using a grinder, the resulting powder is then filtered using a 200 mesh sieve

2.2.2. Carbonization and Activation

Biosorbent from oil palm empty bunches that have been dried is then heated using a furnace with a temperature of 500°C for approximately 2 minutes. After the carbonization process, the biosorbent is allowed to cool naturally. Biosorbents that have been carbonized are prepared and weighed. . A total of 10 grams of carbon was immersed in 100 mL of 10% H3PO4 (1:10) for 24 hours. The activated carbon is filtered using filter paper and a vacuum filter and washed with aquades to neutral pH.[7]

2.2.3. Synthesis of AC-Cu(TAC)$_2$ and AC-Fe(TAC)$_3$

The synthesis composite of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$ carried out in this study using the metals $Cu(NO_3)_2.3H_2O$ and $FeCl_3.6H_2O$ was carried out in two stages, namely immersion of 0.3 grams of activated carbon with 10 grams of ethanol and 0,6 grams of TAC for 24 hours, then the solution was evaporated for 24

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hours until the ethanol had evaporated completely and refluxed for 8 hours with a mixture of 0.9 grams of $Cu(NO_3)_2.3H_2O$ or $FeCl_3.6H_2O$, 0.2 mL HF, 0.19 mL HNO_3 and 20 mL aquades. The composition used uses a 1:3:2 ratio between activated carbon, metals, and ligands

2.2.4. Characterization

Characterization of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$ was performed using SEM-EDX for morphological analysis of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$, XRD instruments to determine the crystallinity level of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$.

3. RESULTS AND DISCUSSION

3.1. X-Ray Diffraction (XRD) Characterization

In determining the structure and degree of crystallinity of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$ *X-Ray Diffraction* (XRD) analysis can be used. The data from XRD analysis of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$ is shown in the figure below :

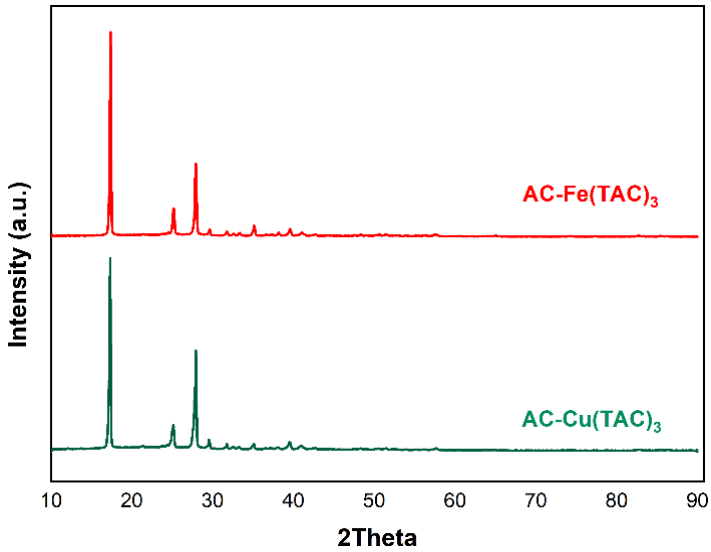

Figure 1. XRD patterns of AC-$Cu(TAC)_2$ and AC- $Fe(TAC)_3$

The results of XRD analysis of AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$ show that the two materials form a crystalline material structure with an XRD pattern that is not much different. AC-$Cu(TAC)_2$ and AC-$Fe(TAC)_3$ show a 2 sharp peaks of diffraction at 2 theta around 15°-30° with a material crystallinity level of 40% by forming a relatively similar crystal system, namely the triclinic crystal system. However, the two materials have different crystal volumes, namely 735.07 Å compared to 235.67 Å. This shows that AC-$Cu(TAC)_2$ has a larger crystal volume compared to AC-$Fe(TAC)_3$. This is supported by the stability of Cu metal-based MOFs

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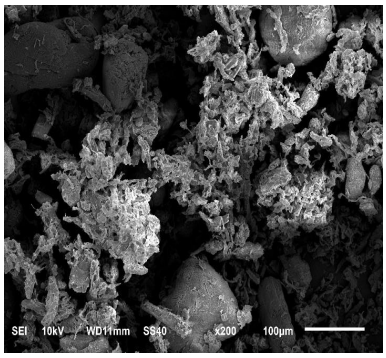
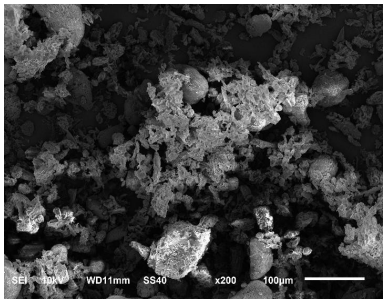
which show a strong interaction between the Cu^{2+} of the metal and the -O- of the organic ligand which makes the material more stable.[8]

3.2. Scanning Electron Microscopy (SEM) Characterization

The morphological analysis of AC-Cu$(TAC)_2$ and AC-Fe$(TAC)_2$ composite uses SEM analysis with 200x magnification to see the surface shape produced by AC-Cu$(TAC)_2$ and AC-Fe$(TAC)_3$.



(A)



(B)

Figure 2. SEM Analysis (A) AC-Cu$(TAC)_2$ and (B) AC-Fe$(TAC)_3$

Based on figure 2(A), at 200x magnification, it can be seen that AC-Cu$(TAC)_2$ forms fibers that are bound to each other and attached to the cross-section, have relatively different sizes but have relatively large porosity so that it shows a fairly strong bond between activated carbon and MOFs Cu(TAC). In figure 2(B) we see a relatively unequal size of the fragmented fiber particles with smaller porosity, this shows a weak bond between activated carbon and MOFs Fe(TAC).

3.3. Energy Dispersion X-Ray (EDX) Characterization

In this study, the components found in AC-Cu$(TAC)_2$ and AC-Fe$(TAC)_3$ are identified using EDX (Energy Dispersion X-ray) analysis. The basic premise behind this technique is that each element has a unique atomic structure, allowing for a different collection of peaks in its electromagnetic emission spectrum. The EDX characterisation shows the weight percentage (Wt%) and atom percentage (At%) for each element in the sample.

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EDX result in Figure 3 shows that oxygen is the highest element in AC-Cu(TAC)$_2$ by weight (Wt%) of 61,33% and percent of atoms (At%) of 56,48%. Other elements indicated in AC-Cu(TAC)$_2$ are Carbon and Cu elements which each have a weight percent (Wt%) of 33,02% and 0.66% and atomic percent (At%) only obtained by C and Cu elements of 40,64% and 0,15%. Figure 4 shows the highest element in AC-Fe(TAC)3 by weight (Wt%) of 52,22% and percent of atoms (At%) of 47,70%. Other elements indicated in AC-Fe(TAC)3 are Carbon and Fe elements which each have a weight percent (Wt%) of 40,69% and 2,68% and atomic percent (At%) only obtained by C and Fe elements of 49,50% and 0,70%.

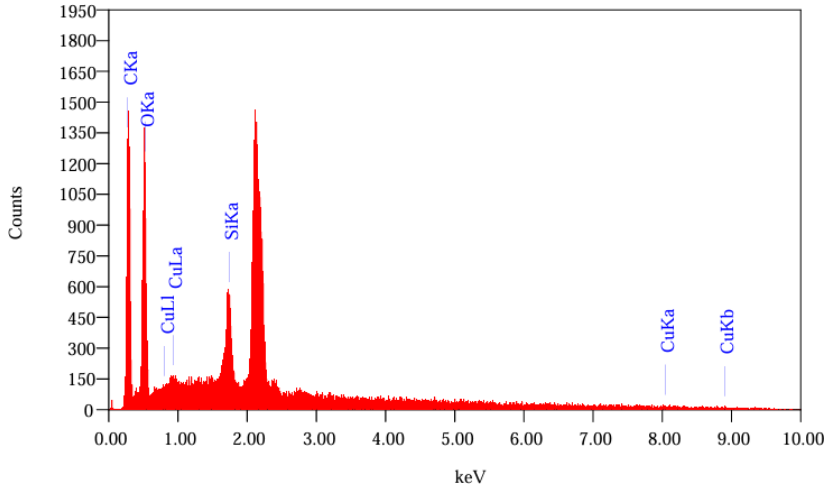

Figure 3. Characterization of EDX AC-Cu(TAC)$_2$

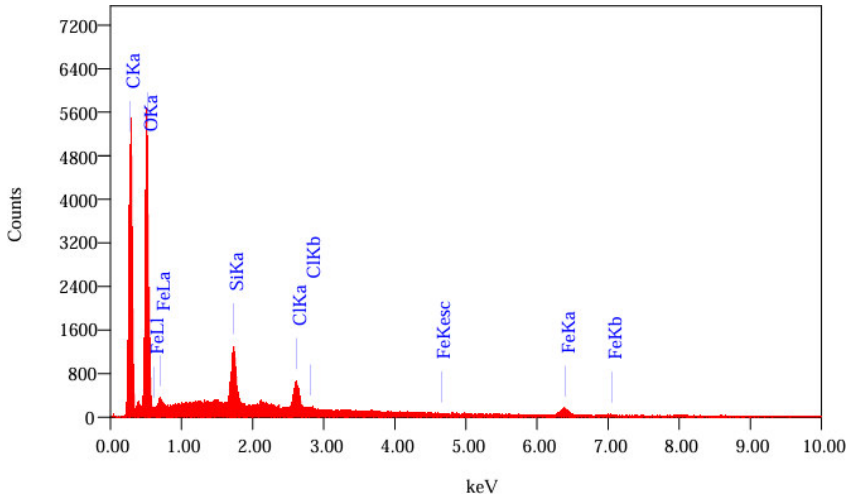

Figure 4. Characterization of EDX AC-Fe(TAC)$_3$

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4. CONCLUSION

Characterization of AC-Cu(TAC)$_2$ and AC-Fe(TAC)$_2$ indicated that the observed XRD pattern is characteristic of the structure of crystalline materials, with AC-Cu(TAC)$_2$ and AC-Fe(TAC)$_3$ having a crystallinity of 40% and forming a triclinic crystal system. A relatively large porosity in AC-Cu(TAC)$_2$ indicates that the fibers are interbonded and attached to the cross-section, suggesting a strong bond between activated carbon and MOFs Cu(TAC). The size of the fragmented fiber particles in AC-Fe(TAC)$_3$ is relatively different from the smaller porosity. These findings are supported by characterization based on SEM at 200x magnification. Cu and Fe metals are identified as composite materials using EDX analysis.

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