The Effect of Temperature in the Application Of Mesoporous Nanomaterials Based on Carbon in Drug Delivery System With Ibuprofen

Maria Ulfa1, Reni Alfi Ardini1, and Didik Prasetyoko2

1 Study Program of Chemistry Education, Faculty of Teacher Training and Education, Universitas Sebelas Maret, Ir. Sutami 36A, 57126 Surakarta, Central Java Indonesia
2 Department of Chemistry, Faculty of Mathematics and Natural Sciences, Institut Teknologi Sebelas November, Jl. Keputih, Surabaya, East Java Indonesia

email : ulfa.maria2015@gmail.com

Abstract: In this research, we present synthesis of high porosity and surface areas of nanomaterials Based on mesoporous carbon (MnbC) with hard tempering route. Firstly, silica hard template route assembly was prepared by using tetraethoxysilane (TEOS) and triblock copolymer Pluronic F127 roomates is employed as directing templating agents under acidic conditions. By using powder silica template and sucrose catalyzed by H2SO4 in the synthesis mixture, MnbC were generated after calcination Followed by organic removal in the common washing process. Character of the MnbC was studied by scanning electron microscopy and surface analysis techniques. The result is the MnbC has structure like a wire, a large pore volume and pore diameter, is inert in an aqueous, acidic and alkaline environments. With such characteristics, Ibuprofen adsorption on MnbC run optimally when released at 45°C and shows the relationship between temperature and adsorption in experiment.

1. Introduction

At this time many people who discussed the nanoparticle technology in relation to the development of drug delivery systems. Particles or globules at the nanometer scale has unique physical properties compared to the particles at a larger size, especially in improving the quality of drug compounds penghantaran[1]. Nanotechnology can be defined as the design, characterization, manufacture and application of a structure or system in a way to control the shape and size at the nanometer scale, while nanomaterials are defined as objects / materials that have one dimension in the nanometer scale (1 - 100 nm )[2]. One form of nanoparticles, namely activated carbon. Based on the pore size of the porous material can be classified into micropores, mesopores and macropores. Where microporous for particles with pore sizes less than 2 nm. Macropore for particles with a pore size above 50 nm. While mesoporous are particles with a pore size between 2 to 50 nm[3]. Based on this it can be stated that the mesoporous carbon is a form of activated carbon nanoparticle technology with pore sizes between 2 to 50 nm.

Mesoporous carbon can be prepared by the method of templating soft or hard templating. Templating method is basically mixing a source of silica to the carbon, then the mixture was heated to form a composite composed of solid carbon and silica. In the final stage, the silica is removed by using a solvent extracted base[4]. Mesoporous carbon used in this study has been prepared with hard templating method using mesoporous silica material SBA-15 with sucrose as a template as well as a carbon source. The use of hard templating technique is done in order to overcome the complexity of
the synthesis stage mesoporous carbon using soft techniques based surfactant templating. By 1997, the technique has not been able to get a soft templating of mesoporous carbon with the regularity of a good structure and porosity. Some of the causes of failure is very difficult to get organized with the involvement of mesoporous carbon surfactant micelles as a template for software vulnerable to damage caused by changes in pH, solvent, chemical reactions (outside the synthesis process), humidity, temperature and time[5].

Ibuprofen (acid 2 - (- 4-isobutylphenyl) propionic acid) is a nonsteroidal anti-inflammatory drugs are often used in the medical world[6]. Ibuprofen is an anti-inflammatory drug with low water solubility but has good permeability in the digestive tract system[6]. Ibuprofen nature which has a low solubility in water poses a problem in the natural environment after going through an industrial process. Many studies that examine in an effort to minimize the level of Ibuprofen in water molecules such as coagulation, adsorption, filtration, and separation. Adsorption is the most efficient technique to remove ibuprofen[5]. Adsorbs to lower levels of Ibuprofen molecule requires optimum conditions, but so far research on the molecular adsorption to lower levels of Ibuprofen rarely focus on the different temperature conditions. The focus in this research is to investigate the adsorption loading of Ibuprofen time at different temperatures.

2. Methods
2.1 Material
Materials used in this study is Pluronic P123 surfactant used as a soft directing agent in the synthesis of SBA-15. Other materials used are HCl, H₂SO₄, NaOH were purchased from Sigma-Aldrich. In addition, also used deionized water and sucrose as a carbon source purchased from Merck for synthesis MNbC.

2.2 Synthesis of SBA-15
The initial step in this experiment is to perform the synthesis of mesoporous silica SBA-15. The first step in this synthesis is to shed P-123 into a solution of HCl at a temperature of 40°C while distirer for 20 hours. The resulting solution is then added tetraethyl orthosilicate (TEOS) dropwise and the mixture was kept in a water bath at 40°C for 20 hours while distirer. The ratio of P-123: HCl: TEOS is 0.020: 0.750: 0048. The resulting mixture was then hydrothermally with a closed container at a temperature of 100°C for 48 hours. The next step is to separate the solid product was washed with water, then dried at a temperature of 60°C, and finally calcined at 550°C in air for 6 hours to remove the soft template P-123. White product is labeled as mesoporous silica SBA-15.

2.3 Synthesis of Mesoporous Carbon
The next procedure is the synthesis MNbC using SBA-15 as hard template. MNbC preparation begins by dissolving the mesoporous silica SBA-15 into the sucrose solution for 60 minutes. Sucrose solution containing sucrose, sulfuric acid and deionized water with a ratio of 0.125: 0.010: 0.500 distirer at 200 rpm for 30 min at room temperature. White composite after completion of mixing heated in an oven at two different temperatures, namely at 110°C and 160°C for 6 hours. Chocolate samples after the heating process was mixed with the sucrose solution. The second sucrose solution containing sucrose, sulfuric acid and deionized water with a ratio of 0.080: 0.005: 0.500. After the mixing process, the sample is prepared by heating both the 110°C and 160°C for 6 hours. Dark chocolate samples hydrolyzed at a temperature of 900°C for 12 hours in a stream of nitrogen gas with a heating rate of 3C / min. At the end of the synthesis process, the removal of SBA-15 silica mesoporous done washing step using sodium hydroxide solution at room temperature followed by the stirrer 30 minutes, filtering, washing and drying at 110°C for one night. Mesoporous carbon produced in this synthesis are labeled MNbC.

2.4 Characterization
In this study, characterization used are Scanning Electron Microscope (SEM) to determine the level of MNbC. In addition to using the data of SEM, the study also using FTIR to determine the structure and morphology as well as UV-Vis spectrophotometer is used to measure the transmittance or absorbance of the sample as a function of wavelength.

2.5 Ibuprofen adsorption on MNbC
Preparation of Ibuprofen molecular adsorption on MNbC made by mixing 10 ml of ibuprofen solution with concentration of 100 ppm onto 0.005 g of powder of MNbC. Then a solution on the stirrer at a temperature of 0°C and then sampled A / 0°C at intervals of sampling at time 0 minutes, 5 minutes, 10 minutes, 15 minutes, 20 minutes, 30 minutes, 35 minutes, 45 minutes and 60 minutes. The samples in the stirrer at different temperatures (37°C and 45°C) expressed as B / 37°C and C / 45°C.

3. Results and Discussion
In Figure 1. A and B show three-dimensional images of structures MNbC after the release of the characterization using scanning electron microscopy (SEM). In the picture shows that MNbC looks like a wire. Wire as in the SEM images showed that the infiltration of the carbon source to SBA-15 as a mold have been successfully synthesized by templating process. Based on the results of characterization SEM images showed that the difference in MNbC condition before and after adsorption using Ibuprofen. After adsorption, the molecular MNbC damaged ± 20% of the total MNbC used in the adsorption process. MNbC molecular damage after adsorption occurs because the stirrer process conducted during the experiment damaging the molecular structure of MNbC.

In. Figure 1. Shows that the blue circle is MNbC which looks like a wire, while the one in the red circle shows MNbC which is damaged and covered by Ibuprofen. Based on two image characterization using scanning electron microscopy (SEM) after adsorption process, it appears that there is a lot of residual Ibuprofen molecule which is not attached MNbC. It shows that the adsorption process that lasts less than the maximum, so that Ibuprofen is supposed adsorbed into the pore MNbC can not be adsorbed completely. This happens because MNbC have pores that are not the same so some Ibuprofen molecule with a larger size can not be adsorbed.

FTIR analysis is used to determine the functional groups contained in the solution after adsorption. Activated carbon has functional group uptake on the surface of the activated carbon, such as functional
groups C-O, -C=O, and O-H. Vibration C-O appearing at wavelengths 1019 cm$^{-1}$, and vibration O-H appears at 3435 cm$^{-1}$ and 3677 cm$^{-1}$ with wide arches (See Figure 2).

![Figure 2. Characterization FTIR of MNbC after adsorption](image)

Ibuprofen in the infrared shows the peak in 1721 cm$^{-1}$, 1232 cm$^{-1}$, 779 cm$^{-1}$, 1185 cm$^{-1}$, 1273 cm$^{-1}$ and 870 cm$^{-1}$[7], Ibuprofen is typical of the group showed a vibration C-H, C=O and C-C. In figure 2 C-H vibration appears at wave number 2915 cm$^{-1}$ and 2750 cm$^{-1}$. Briefly, the results of FTIR characterization can be presented in the following table1.

| type bond | wavenumber |
|-----------|------------|
| C – O     | 1019 cm$^{-1}$ |
| O – H     | 3435 cm$^{-1}$ |
|           | 3677 cm$^{-1}$ |
| C - H     | 2915 cm$^{-1}$ |
|           | 2750 cm$^{-1}$ |

Figure 3. indicate that maximum adsorption occurs at a temperature of 45˚C and declines as the lower temperature is the temperature experiment 37˚C and 0˚C. This is due to the relationship between the temperature of the surface tension. The process of adsorption increases with decreasing surface tension[8]. Thus, the adsorption process that takes place will increase with decreasing surface tension due to the increase in temperature. At a low surface tension, the greater the surface area of the adsorbent adsorbate which interacts with the interaction between the adsorbate with an adsorbent becomes larger and the adsorption process will increase.

Adsorption capacity in this study were calculated using the Langmuir equation[9].

\[
\ln(q_e - q_t) = \ln q_e - k_1t
\]

\[
\frac{t}{q_t} = \frac{1}{k_2(q_e)^2} - \frac{1}{q_e} t
\]

\[
q_e = q_{max} \frac{k_L q_e}{1 + k_L q_e}
\]
\[ \frac{c_e}{q_e} = \frac{1}{q_{max} K_L} + \frac{C_e}{q_{max}} \] (4)

Based on calculations, showed that q_max at temperature 0°C is = 1408.45 mg/g; 37°C is = 740.74 mg/g; 45°C is = 1063.82 mg/g. Gibbs free energy change (ΔG°) Can be calculated using the Langmuir constants are included in the equation (5)[10].

\[ \Delta G^\circ = -RT \ln K_L \] (5)

Calculation resulting value ΔG° = -7071.19 kJ/mol which indicates that the adsorption process takes place spontaneously and produce energy bond between Ibuprofen with relatively low MNbC (<40 kJ / mol) so that it can be concluded that the adsorption takes place in the category of physical bonding.

![Figure 3. The adsorption of Ibuprofen on MNbC at a temperature of a. 45°C, b. 37°C, and c. 0°C](image)

4. Conclusions
MNbC can be synthesized using the hard templating technique route and produce carbon which looks like a wire. Ibuprofen adsorption on MNbC run optimally when released at 0°C and shows the relationship between temperature and adsorption in experiment. The lower adsorption temperature, the higher the ability of MNbC to adsorb Ibuprofen. The adsorption process takes place spontaneously and produce energy bond between Ibuprofen with relatively low and category of physical bonding.

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