Indonesian Journal of Chemical Research
http://ojs3.unpatti.ac.id/index.php/ijcr Indo. J. Chem. Res., 8(3), 242-246, 2021

Mesoporous Silica MCM-48 as Chloramphenicol Adsorbent

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Received: November 2020
Received in revised: November 2020
Accepted: December 2020
Available online: January 2021

Abstract

This study is aimed to determine the potential use of MCM-48 to adsorb chloramphenicol pollution. Chloramphenicol adsorption was conducted at various times contact and concentration. Adsorption isotherm was studied by comparing Langmuir and Freundlich adsorption isotherm. The results showed that chloramphenicol adsorption was suited to Freundlich isotherm with an optimum contact time of 80 minutes

Keywords: MCM-48, antibiotic, chloramphencol, adsorption capacity

INTRODUCTION

Before people discover antibiotics, infectious diseases were very deadly for humans (da Chuha et al., 2019). After Flemming discovered penicillin in 1928, antibiotics were produced on a commercial scale then continued to expand. ANtibiotics is not only for medicinal purpose but also use in animal husbandry and the agricultural industry (cdc.gov, 2019). The widespread use of antibiotics is not without consequences. Bacteria that can be inhibited by certain antibiotics quickly adapt and develop immunity and become resistant to these antibiotics (Heinemann, 1999; Lee Ventola, 2015; Michael et al., 2014).

The costs of antibiotic resistance (RA) are currently enormous. The World Health Organization (WHO) estimates that until 2017 there were 700,000 deaths due to antibiotic resistance worldwide and predicts that in 2050 there will be 10 million deaths per year (WHO.int, 2019). The increased risk of RA does not reduce global antibiotic consumption. The antibiotics production and consumption on a large scale can initiate the antibiotic release into the environment. The study found traces of antibiotic contamination in estuaries, surface freshwater even drinking water (Zheng et al., 2011; Danner et al., 2019; Ye and Weinberg, 2007). Therefore, the prevention of antibiotic contamination is needed.

The adsorption method is an effective and inexpensive method that use to tackle pollutants in liquid waste. Researches methods for treating pharmaceutical waste, such as anaerobic processes (Chelliapan et al., 2011; Larsson et al., 2007), chemical oxidation, and physicochemical techniques have been carried done, but have not yielded satisfactory results (Oktem et al., 2008; Hey, 2013; Jiang and Zhou, 2013). The adsorption method with mesoporous silica is an alternative solution in treating liquid waste containing antibiotics. One type of mesoporous silica is MCM-48. The MCM-48 has a high surface area, regular pores with a narrow pore size distribution, high pore volume, and has high biocompatibility (Taba et al., 2017; Gau et al., 2017; Nairi et al., 2017; Taba et al., 2018). These properties make it possible to use it as an adsorbent for antibiotic molecules.

One of the antibiotics often used is chloramphenicol. In this study, the adsorption of chloramphenicol compounds with MCM-48 was carried out to study the potential of MCM-48 as a potential antibiotic waste adsorbent.

METHODOLOGY

Materials and Instrumentals

The tools used were an oven (Type Spini SOSFD), analytical balance (Ohauss), magnetic stirrer (Fisher Type 115), hotplate stirrer (Ikari Basic 1), vacuum pump (Type ME4C), UV-Vis spectrophotometer (Spektronik 20 D+), Prestige-21 FT-IR spectrophotometer, and X-ray diffractometer (Bruker D2 Phaser 2ndGen). The materials used in this study were obtained from Merck and Sigma-Aldrich with high purity. The ingredients are Ludox HS-40, Cetyltrimethylammonium Bromide (CTAB), Triton X-100, absolute ethanol (C_2H_5OH), sodium hydroxide (NaOH), acetic acid (CH_3COOH), chloramphenicol ($C_{11}H_{12}C_{l2}N_2O_5$), and distilled water (H_2O).

Synthesis of MCM-48

The MCM-48 material was synthesized using the Ryoo (1999) method modified by Taba (2001). MCM-

48 was prepared by mixing a tetracilicate solution (14.3g Ludox HS-40 and 45.25 g of 1M NaOH solution) with a surfactant solution (6.12 g CTAB; 1.34 g Triton X-100; and 83.47 g distilled water) in in a propylene bottle then shake for 15 minutes. The mixture was heated at 100oC for 24 hours while shake then cooled to room temperature. The pH was adjusted to 10 by using 30% acetic acid. The mixture was then heated at 100 °C for 24 hours then cooled to room temperature. The MCM-48 formed was filtered, washed with distilled water, and then dried in an oven at a temperature of 120 °C.

MCM-48 characterization

The synthesized MCM-48 result was characterized by the FT-IR method to determine the functional groups. The X-Ray Diffraction was carried out to see the crystallinity.

Chloramphenicol Adsorption

The adsorption of chloramphenicol with MCM-48 was carried out to determine the optimum concentration, time, and adsorption kinetics. In order to determine the optimum adsorption time, 0.2 g of product MCM-48 was used to adsorb chloramphenicol in a solution with a concentration of 20 ppm in the period of 5-120 minutes until the optimum adsorption time was obtained. In determining the optimum concentration of adsorption, 0.2 g of MCM-48 was used to absorb chloramphenicol in solution at several concentrations during the optimum time. The chloramphenicol concentrations before and after adsorption were measured using a UV-Vis spectrophotometer.

RESULTS AND DISCUSSION

Characterization using X-ray diffractogram (XRD)

The X-ray diffraction pattern of MCM-48 in this study (Figure 1) shows that MCM-48 is amorphous. The pattern obtained is not optimal, because of the limited tools used.

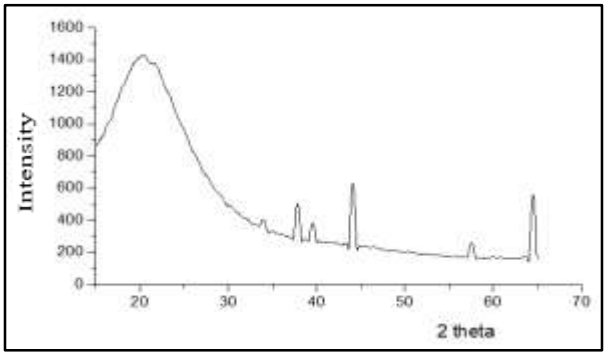

Figure 1. X-ray diffraction pattern of the synthesized MCM-48

The X-ray diffraction pattern of MCM-48 in this study was measured from $2\theta = 15^0$, while the typical MCM-48 pattern was at a small 2θ angle (below 10^0).

Fourier Transform Infra-Red Spectrometer

The results of characterization by FT-IR (Figure 2) showed that MCM-48 contained -OH, -CH, and Si-O functional groups. The organic groups -CH range are symmetrical and asymmetrical at wavelengths 2852.72 and 2922.16 cm^{-1}. The -CH groups are bent at wave number 1355.96; 1415.75; and 1471.69 cm^{-1} are surfactant groups that are still present in the material. The observed wave numbers 1060.86 and 1226.73 cm^{-1} are the symmetrical and asymmetrical stretches of the silicate lattice of the material. The Si-O stretching vibration of Si-OH was observed at wave number 962.48 cm^{-1}. The data obtained is similar to the data in Sari et al's (2015) study, so this data can be confirmed that the material being synthesized is MCM-48 mesoporous silica.

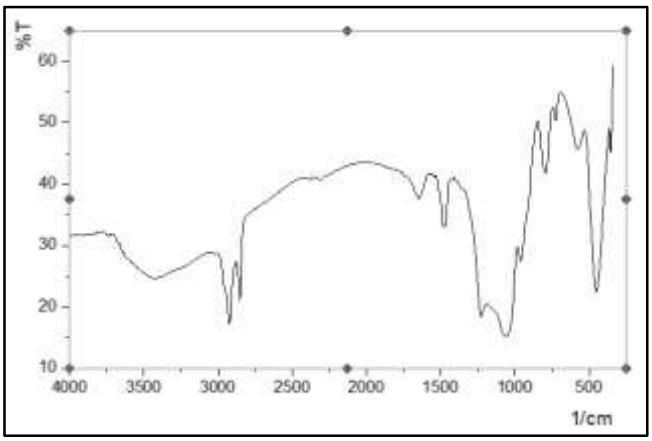

Figure 2. FTIR spectra of synthesized MCM-48

Surface Area Determination by BET Method

Isothermal adsorption and desorption of N2 gas on MCM-48 before washing can be seen in Figure 3. The adsorption isotherm by MCM-48 without washing corresponds to the type IV isotherm. Capillary condensation occurs in the adsorption process indicated by the presence of hysteresis in the N2 gas adsorption-desorption curve. The surface area (BET) of MCM-48 is 851,797 m^2/g. The pore surface distribution shows in Figure 4.

The pore radius based on the BJH method on MCM-48 was 15.268 Å. These results indicate that the synthesized MCM-48 is a mesoporous material.

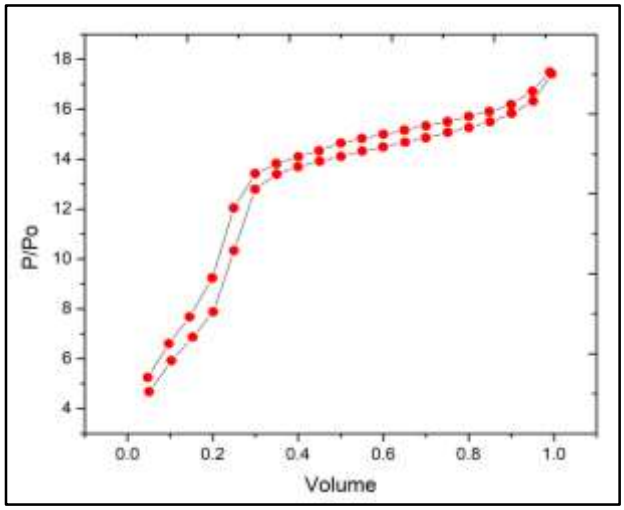

Figure 3. Isothermal N_2 adsorption of MCM-48 before washing

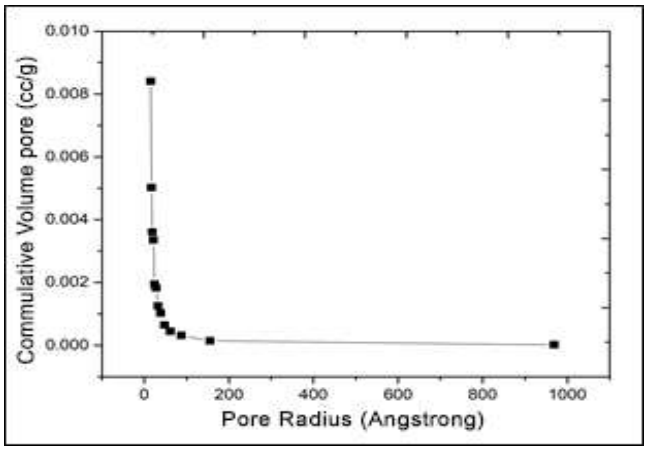

Figure 4. Pore size distribution of MCM-48

Chloramphenicol adsorption by MCM-48

The optimum time of chloramphenicol adsorption by MCM-48

The optimum adsorption of chloramphenicol with MCM-48 was at 80 minutes contact time. Figure 5 shows that the number of chloramphenicol molecules absorbed in MCM-48 tends to be the same at 10 to 45 minutes until finally reaching its peak at 80 minutes. After 120 minutes, the number of chloramphenicol molecules absorbed decreased, which indicates that the adsorbent surface was already saturated.

Study of Chloramphenicol Adsorption Kinetics by MCM-48

By plotting the t/q_e value against contact time (Figure 7), the R^2 value is 0.9998. Thus, chloramphenicol adsorption on MCM-48 is pseudo-second-order adsorption.

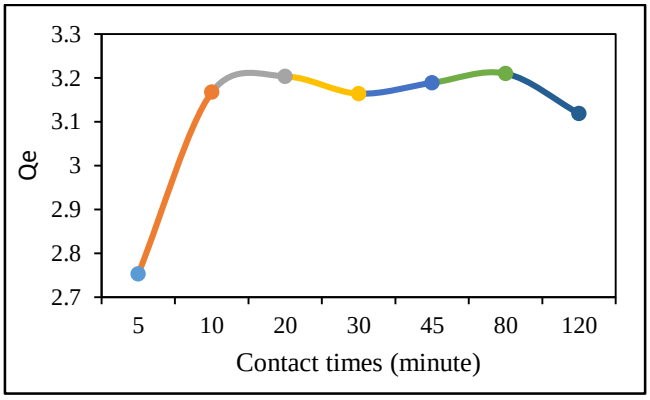

Figure 5. The number of chloramphenicol molecules adsorbed (q_e) by MCM-48 as a function of contact time

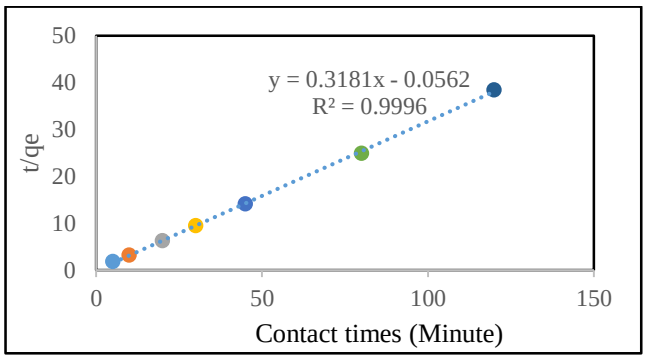

Figure 6. Chloramphenicol adsorption order on MCM-48

Adsorption Capacity of chloramphenicol by Mesoporous Silica MCM-48

The absorption of chloramphenicol molecules tends to increase with increasing number of molecules in the same volume (Figure 7). Using this data, the adsorption capacity is determined based on the Langmuir and Freundlich isothermal model. Langmuir isothermal model was created by connecting the linear curve C_e/q_e to C_e, while the Freundlich isothermal model was created by connecting the linear log q_e curve to log C_e. The second isothermal model, both Langmuir and Freunlich is shown in Figures 8 and 9.

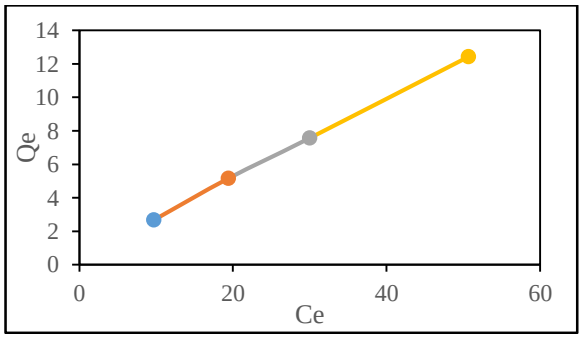

Figure 7. Number of chloramphenicol adsorbed (q_e) by MCM-48 at various concentrations (C_e)

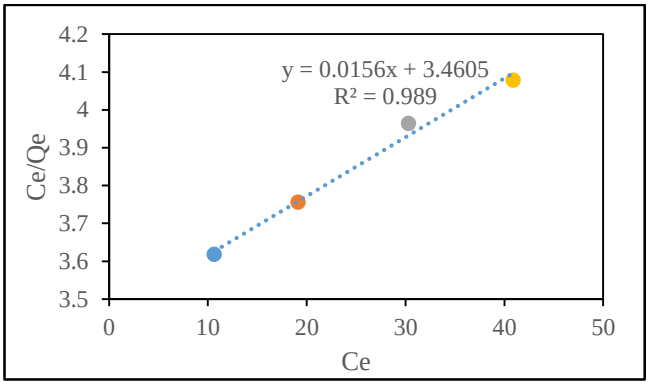

Figure 8. Graph of Langmuir isothermal model

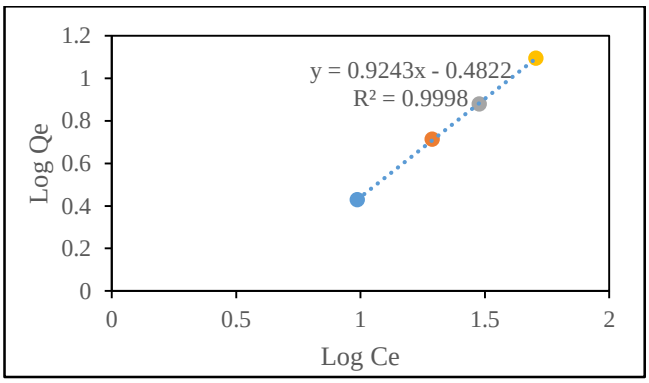

Figure 9. Graph of Freundlich's isothermal model

From these curves, it can be observed that the R^2 value in the Freundlich isothermal model is greater than the R^2 value in the Langmuir isothermal model. Therefore, chloramphenicol adsorption using MCM-48 tends to follow Freundlich's isotherm. The Freundlich equation assumes that adsorption occurs multi-layer on the surface of the adsorbent and adsorbent. Therefore, the adsorption of chloramphenicol with MCM-48 occurs physically.

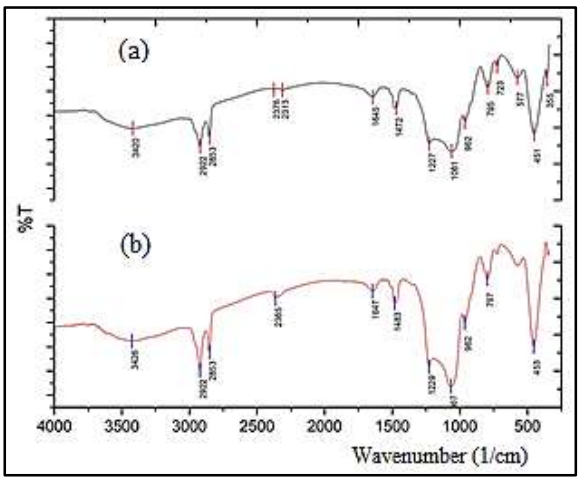

Figure 10. FTIR spectra (a) before adsorption and (b) after adsorption

CONCLUSION

Adsorption of chloramphenicol antibiotic compound with optimum MCM-48 at 80 minutes according to Freundlich's adsorption isotherm. The adsorption of these compounds occurs physically with pseudo-order II. CMM-48 has the potential to be applied as an absorbent material for chloramphenicol in waste.

ACKNOWLEDGMENT

The authors would like to give thank you for the Master's research obtained from the Minister of Research and Technology

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