MESOPOROUS SILICA MODIFIED WITH AMINO GROUP (NH₂-MCM-48) AS ADSORBENT OF Ag(I) AND Cr(III) IN WATER

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ABSTRACT

The amino-modified mesoporous silica (NH₂-MCM-48) was synthesized using a mixture of surfactants as a template that was re-extracted using a methanol-hydrochloride solution. The modifying agent was 3-aminopropyltrimethoxysilane (3-APTMS). The characterization was conducted using an X-ray diffractometer (XRD), Fourier Transform Infrared (FTIR) spectroscopy, and Scanning Electron Microscopy (SEM). We investigated the reduction of Ag(I) and Cr(III) concentrations in the solutions using the modified material under several variables, i.e., pH, contact time, and initial ion concentration. An atomic absorption spectrophotometer measured the concentrations of the ions. The XRD and FTIR results proved that the basic material was MCM-48, and the template was mostly removed after the extraction, respectively. The modified material was successfully produced showed by the FTIR peak of N-H stretching vibration. The equilibrium of the Cr(III) adsorption was achieved in a longer time than that of the Ag(I) adsorption. The adsorption kinetics of Ag(I) and Cr(III) ions obeyed a model of pseudo-second-order and followed better the Langmuir adsorption model than the Freundlich, with a capacity of 0.87 and 2.02 mmol g⁻¹, respectively. The amine-modified mesoporous silica, NH₂-MCM-48 for this study is a very high potential as an adsorbent of metal ions used in the study.

Keywords: Adsorption, 3-APTMS, Mesoporous Silica, Heavy Metals.

INTRODUCTION

Silver (Ag) and chromium (Cr) are heavy metals that can cause health problems to live organisms as well as damage the habitat and water ecosystem. Chromium (III) found in water can be converted to chromium (VI) which is toxic.¹,² If it accumulates in the body, it will cause cancer and genetic transmutation. For these reasons, it is necessary to reduce their concentration in the water environment. Several techniques have been reported for separating heavy metal ions from wastewater, such as ion exchange,³ precipitation,⁴ membrane technology,⁵,⁶, and adsorption.⁷-¹⁶ Adsorption is a method widely used because it is simple and effective.¹⁷ The adsorbents used can derive from natural sources or synthetic materials. The synthetic ones can be porous or non-porous materials. Porous materials are divided into microporous, mesoporous, and macroporous. Mesoporous substances are interesting to be used as adsorbents because they have high surface areas and large pore volumes.

One of the mesoporous materials is mesoporous silica (MCM-48), which belongs to an M41S family discovered by researchers from Mobil Oil Corporation. They are amorphous with well-defined pore shapes and sizes, have large specific surface areas and specific pore volumes.¹⁸,¹⁹ The mesoporous material, MCM-48, is more attractive as an adsorbent due to the cubic structure that produces three-dimensional channels protecting the blockage of the pores by guest molecules and making it easier for the diffusion of molecules into the pores.²⁰ Silanol and siloxane groups are usually found in the substance. They can interact with heavy metal ions. To increase the ability in adsorbing metal ions, modification of the surface of MCM-48 is a promising method because several modifying agents are available. One of the functional materials that are most frequently used to modify the kind of silica is 3-aminopropyltrimethoxysilane (3-APTMS).

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The current research of the modified silica has been utilized effectively as an adsorbent of heavy metal ions from aqueous solutions. Amino (-NH$_2$) group in 3-APTMS is a ligand of the hard base that can strongly interact with a hard acid such as Cr(III) ion. However, a previous study showed that the amino-functionalized MCM-48 interacted well with a borderline acid, Cu(II) ion. In this work, the adsorption of the soft acid, Ag(I) ion, on the amine-functionalized MCM-48 was conducted to compare the interaction of the material with other kinds of acids. As a comparison, the Cr(III) ion was also used as an adsorbate. Adsorption processes depend on pH, contact time, and concentration; therefore, the adsorptions of Ag(I) and Cr(III) ions were studied under various pH, contact time, and initial concentration. Results showed that NH$_2$-MCM-48 was a very promising adsorbent of the studied ions.

**Experimental**

**Material**
The materials used were cetyltrimethylammonium bromide (CTAB), Ludox HS-40 purchased from Du Pont, Triton X-100, 3-aminopropyltrimethoxysilane (3-APTMS) from Sigma-Aldrich, base (NaOH), acids (HCl and CH$_3$COOH) as well as AgNO$_3$ and Cr(NO$_3$)$_3$·9H$_2$O as the heavy metal sources. They were used directly used without any treatment.

**Synthesis of MCM-48 and Amine-modified MCM-48**
The synthesis of MCM-48 and the removal of surfactant from the as-synthesized MCM-48 followed the procedure mentioned in the previous study without the addition of NaCl. The XRD patterns of materials before and after the removal of surfactant (template) were obtained by using a diffractometer (Siemens D500), using CuK$_\alpha$ as a source of radiation with a wavenumber of 1.5412 Å. Samples were placed into an aluminum holder and the diffraction patterns were created in a continuous scan mode (a scan rate = 1 degree 2-theta/min and a step size = 0.02 degree 2-theta). The divergence slit was 0.3 ° and the receiving slit was 0.05 °. The experiments were conducted at a current of 30 mA, an accelerating voltage of 40 kV, and a scan range from 0.8 to 10 degree 2-theta. The FTIR spectra of samples were obtained using a Shimadzu (IR Prestige-21 FTIR Spectrometer) run on 340-4500 cm$^{-1}$ at a resolution of 4, and a scan number of 300. Modification of MCM-48 was conducted following the previous method with some adjustment as has been described elsewhere.

**Experiments of Adsorption Kinetics**
Adsorption kinetic experiments of Ag(I) and Cr(III) ions by NH$_2$-MCM-48 were separately studied using silver nitrate and chromium(III) nitrate solutions with the same initial concentrations of Ag(I) and Cr(III) ions (100 mg/L). The adsorbent used was 100 mg added to 50 mL of the ion solutions and stirred at ambient temperature for times ranging from 10 to 90 minutes for Ag(I) ion and 360 to 1260 minutes for Cr(III) ion on a magnetic stirrer (stirring rate was similar). Filtering removes the solids from the solution, and an atomic absorption spectrophotometer examined the filtrates containing the ions. These experiments can also obtain the optimum contact times of Ag(I) and Cr(III) ions. Experiments without any adsorbent were conducted as control experiments. The pseudo-first-order or pseudo-second-order kinetics were used extensively to describe the adsorption kinetics used. The pseudo-first-order model can be seen in equation (1), derived by Azizian.

$$\ln \left( q_e - q_t \right) = \ln q_e - k_1t$$

(1)

The numbers of ions adsorbed at equilibrium and at time $t$ are $q_e$ and $q_t$, respectively and the rate constant of the pseudo-first-order is $k_1$.

The pseudo-second-order process is shown in equation (2),

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$$

(2)

Where $q_e$ and $q_t$ are the numbers of ions adsorbed at equilibrium and at time $t$, respectively and $k_2$ is the rate constant of the pseudo-second-order process.

**Adsorption of Ag(I) and Cr(III) Ions As A Function of pH**
The adsorption of Ag(I) and Cr(III) ions at various initial pH (2-7) was conducted to study the influence of pH on the adsorption process. Solutions used to adjust the initial pH were HCl and NaOH. The pH adjustment was carried out before the addition of the adsorbent. The experiments were performed at the optimum contact time and the blank experiments without the addition of adsorbent at each pH value were also conducted.
Adsorption of Ag(I) and Cr(III) Ions As A Function of the Initial Concentration of Ions

The adsorption of Ag(I) and Cr(III) ions at various initial concentrations was performed to investigate the effect of concentration on adsorption. The procedure was conducted by mixing the ion solutions with NH$_2$-MCM-48 (100 mg) in a beaker containing 50 mL of a single metal ion solution, carried out at the optimum contact time and pH. The experiments were performed at a concentration range of 100-800 mg/L.

RESULTS AND DISCUSSION

Characterization of MCM-48 Materials

The X-ray diffraction patterns of MCM-48 before and after removing surfactants are given in Fig.-1. The XRD pattern (Fig.-1a) shows the presence of 2-theta with high intensities at 2.21° and 2.44° with the Müller indexes of [211] and [220], respectively. Several peaks; [220], [321], [400], [420], [322], [422], and [321] are observed with low intensities. The peaks are specific of material with the good order of MCM-48 as described in previous works. After the removal of the template, all peaks shift to higher 2-theta due to constriction and condensation of the pores. The intensities of peaks increase after the removal of surfactants because of the absence of scattering caused by the template.

![Fig.-1: XRD Patterns of MCM-48 (a) Before and (b) After Removal of the Template](attachment:image.png)

Functional groups found in MCM-48 before and after extraction of surfactants as well as after modification (NH$_2$-MCM-48) have been reported in the previous study. After the removal of the surfactant from MCM-48, almost all surfactants have been removed. After modification, there was an interaction between silanol groups and 3-APTMS shown by the presence of -C-H, -N-H, -C-N, and Si-CH$_2$-R obtained from 3-APTMS. The FTIR spectra of modified MCM-48 with and without Ag(I) and Cr(III) ions are given in Fig.-2.

![Fig.-2: FTIR Spectra of (a) NH$_2$-MCM-48, (b) NH$_2$-MCM-48 + Ag(I) and c) NH$_2$-MCM-48 + Cr(III)](attachment:image.png)

The peaks at 3432 and 3368 cm$^{-1}$ associated with the stretching –N-H vibration are unified and shift to 3443 and 3445 cm$^{-1}$ after adsorption of Ag(I) and Cr(III) ions, respectively. This finding showed that the ions interacted with the –NH$_2$ group on the surface of the modified MCM-48. The fact is supported by the decrease in the –N-H bending intensity at 1566 cm$^{-1}$ caused by the interaction of ions with –NH$_2$.
groups through electron-pair donors from the nitrogen atom in the amino group. The absorption peak of the vibration also shifts to 1556 cm\(^{-1}\) after the interaction with Ag(I) and Cr(III) ions. Also, the intensity of the absorption band at a wavenumber of 1385 cm\(^{-1}\) increases because the interaction of the ions with the –NH\(_2\) groups can cause the increase of the polarity of NH\(_2\)-MCM-48.\(^{11}\) The SEM images of NH\(_2\)-MCM-48 before and after adsorption of Cr(III) and Ag(I) ions are given in Fig.-3.

The SEM images show that NH\(_2\)-MCM-48 consist of particles ranging from several to dozens of micrometers, and gives an irregular shape with a relatively smooth surface. After adsorption of Ag(I) and Cr(III) ions, there is agglomeration on the surface of the adsorbent caused by the metal ions adsorbed.

**The Kinetic Study and the Adsorption Optimum Time**

The adsorption of Ag(I) and Cr(III) ions on NH\(_2\)-MCM-48 as a function of the contact time is given in Fig.-4. The adsorption of ions increases with increasing contact time. The maximum adsorbed amount is achieved when the equilibrium is reached at the optimum time. The optimum times for the adsorption of Ag(I) and Cr(III) ions are 20 and 900 min, respectively. The contact times were applied for experiments at various pH and concentrations.

![Fig.-3: SEM Images of NH\(_2\)-MCM-48 (a) Before Adsorption, (b) After Adsorption of Ag(I) Ion and (c) After Adsorption of Cr(III) Ion](image)

The kinetic data can be obtained from the amount of ions adsorbed at various contact times as given in Table 1. The correlation coefficients using the first-pseudo-order equation are far from 1. However, the correlation coefficients using the second-pseudo-order equation are close to one. Also, the amount of ions adsorbed at the equilibrium obtained from the second-order equation is closer to that obtained from experiments. The results indicate that the adsorption of Ag(I) and Cr(III) ions are in parallel with the pseudo-second-order equation. The same results were also reported by other studies.\(^{11,26}\) The rate constants \((k_c)\) of Ag(I) and Cr(III) adsorption in this study are \(1.0 \times 10^{-24}\) and \(1.0 \times 10^{-3}\) g mg min\(^{-1}\), respectively. These results are following the results of the optimum contact time, where the optimum contact time for the adsorption of Ag (I) is lower than the adsorption of Cr (III).

![Fig.-4: Effect of Contact Time on Quantity (a) Ag(I) and (b) Cr(III) Ions Adsorbed on NH\(_2\)-MCM-48](image)

| Table-1: Kinetic Data of Ag(I) and Cr(III) Ions adsorbed on MCM-48- NH\(_2\) |
|-----------------|--------|---------|
| Data            | Ag(I)  | Cr(III) |
| Pseudo-first-order | \(k_f\) (min\(^{-1}\)) | 0.01     | 0.001   |
from Lewis acids such as Cr(III) will interact well with Lewis bases such as \( \text{NH}_4^+ \) is higher than that of the later. This finding is consistent with the HSAB principle. According to this principle, hard Lewis acids such as Cr(III) > Ag(I) > Zn(II) > Cu(II).

To determine the adsorption capacity, Langmuir and Freundlich's isotherms were used as shown in Fig. 5. Table 2 shows that the adsorption of Ag(I) and Cr(III) ions fit better Langmuir isotherm with the R² value that closer to one. Some researchers also reported the same result. For Cr(III) ion, the amount adsorbed increases from pH 4 to 5 and then from pH 5 to 7, the amount adsorbed decreases gradually. Thus, the adsorption of Cr(III) achieves a maximum at a pH of 5. The same result was also reported elsewhere.

The low adsorption of ions on the adsorbent at low pH is because of the higher concentration of \( \text{H}^+ \) in the solution that competes with the ions for the adsorption sites. Also, the protonation of the surface of NH₂-MCM-48 to form \( \text{H}_2\text{N}^-\text{MCM}-48 \) can inhibit the ability of the functional groups of the adsorbents to interact with the ions. At higher pH, the amount of ions adsorbed is higher because the protonated surface is lower, causing the higher negative charge on the surface, to enhance the adsorption. Further increasing of pH (at very high pH), precipitation of metal hydroxide occurred resulting in the lower concentration of ions detected in solutions. From the results, it is clear that the optimum pH of Ag(I) and Cr(III) ions are 4 and 5, respectively. The optimum pH was used for studying the effect of concentrations to find the adsorption capacity.

**Adsorption Isotherms**

The adsorption of Ag(I) and Cr(III) ions is enhanced by increasing the initial concentration of the ions solutions. To determine the adsorption capacity, Langmuir and Freundlich's isotherms were used as can be seen in Fig. 6. Table 2 shows that the adsorption of Ag(I) and Cr(III) ions fit better Langmuir isotherm with the R² value that closer to one. Some researchers also reported the same result. For comparison, data from a previous study are also given in Table 2.

Table 2 shows the ions adsorbed on the modified material are in the order of Cr(III)>Ag(I)>Zn(II)>Cu(II). The adsorption capacity (\( q_e \)) obtained from the Langmuir equation was 2.02 mmol g⁻¹ for Cr(III) ion and 0.87 mmol g⁻¹ for Ag(I) ion. The adsorption capacity of the former ion is higher than that of the later. This finding is consistent with the HSAB principle. According to this principle, hard Lewis acids such as Cr(III) will interact well with hard Lewis bases such as \(-\text{NH}_2\) group from 3-APTMS. The previous studies used borderline Lewis acids, Cu(II) and Zn(II).
The findings in this study indicate that a sulfhydryl group which is a soft base. The base can interact well with a soft acid, such as Ag(I) ion. Used for the silica functionalized by other adsorbents used by different researchers, except for the adsorption of Cr(III) ions. It can be seen that the adsorption capacity of Ag(I) ion compared to the borderline Lewis acid is likely caused by the higher the ionic radius of Ag(I) (126 pm) than that of the two ions (Zn(II) = 74 pm and Cu(II) = 73 pm). According to the previous study, the amount of ions adsorbed is directly proportional to the ionic radius. Table-3 shows the comparison of the adsorption capacity of ions used in this study and the ones from other researchers.

| Ions                   | Q₀ (mg g⁻¹) | b | R² | Kf (mmol g⁻¹) | n | R² |
|------------------------|-------------|---|----|--------------|---|----|
| Cr(III)                | 105.26      | 2.02 | 0.14 | 35.12 | 0.69 | 4.79 | 0.45 |
| Ag(I)                  | 93.43       | 0.87 | 0.12 | 25.26 | 0.23 | 4.17 | 0.68 |
| Cu(II)                 | 25.45       | 0.43 | 0.07 | 11.89 | 0.20 | 8.79 | 0.94 |
| Zn(II)                 | 35.71       | 0.56 | 0.03 | 5.68  | 0.09 | 3.18 | 0.98 |

The higher adsorption capacity of Ag(I) ion compared to the borderline Lewis acid is likely caused by the higher the ionic radius of Ag(I) (126 pm) than that of the two ions (Zn(II) = 74 pm and Cu(II) = 73 pm). According to the previous study, the amount of ions adsorbed is directly proportional to the ionic radius. Table-3 shows the comparison of the adsorption capacity of ions used in this study and the ones from other researchers.

| Adsorbents                               | Adsorption Capacity (mg/g) | Optimum pH |
|------------------------------------------|----------------------------|------------|
| Cr(III) ionic imprinting PVA/SA          | 59.91                      | 6          |
| Jackfruit peel modified by EDTA          | 41.67                      | 5          |
| Amino functionalized mesoporous nanofiber membrane | 97                       | -          |
| Fe-doped Biomass-derived ordered mesoporous carbon | 46                       | 6          |
| Mesoporous silica functionalized with 3-mercaptopropyl trimethoxysilane | 114.9                   | 4          |
| Chitosan/polydopamine@C@magnetic fly ash | 57.02                     | 4          |
| Poly(vinyl alcohol) modified with thiourea (TU–PVA) | 66.93                   | 5          |
| Amine-functionalized mesoporous silica (this study) | 105.26                  | 5          | 4          |

It can be seen that the adsorption capacity of the Cr(III) ion on NH₂-MCM-48 is higher than that on other adsorbents used by different researchers, except for the adsorption of the Ag(I) ion on mesoporous silica functionalized by 3-mercaptopropyltrimethoxysilane. This occurred because the modified agents used for the formation of the two materials were different. The modified agent used in Ref.²⁹ contains a sulfhydryl group which is a soft base. The base can interact well with a soft acid, such as Ag(I) ion. The findings in this study indicate that this material is a very promising adsorbents of metal ions.
CONCLUSION

The study concluded that the amount of Ag(I) and Cr(III) ions adsorbed on NH$_2$-MCM-48 achieved a maximum at the contact time of 20 and 900 min and pH of 4 and 5, respectively. Both ion adsorptions fitted the pseudo-second-order in which the rate constant of Ag(I) adsorption was higher than Cr(III) adsorption. Both ion adsorptions followed the Langmuir isotherm. The adsorption capacity of the Cr(III) ion (2.02 mmol g$^{-1}$) was higher than that of the Ag(I) ion (0.87 mmol g$^{-1}$). FTIR data clearly showed that there was an interaction between the ions and the functional group of the adsorbent. The interaction of both ions with NH$_2$-MCM-48 caused the shift of the stretching vibration of –N-H and the decrease of the bending –N-H intensity as well as the increase of the –C-N intensity.

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