

DOI: <https://doi.org/10.24425/amm.2023.145478>M. IBRAHIM^{1,2}, W.M.W. IBRAHIM^{2,3}, M.M. AL B. ABDULLAH^{2,1},
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THE INFLUENCE OF FOAMING AGENT TOWARDS METAKAOLIN BASED ALKALI ACTIVATED MATERIALS PROPERTIES AND Cu²⁺ ADSORPTION

The performance of adsorbent synthesized by alkali activation of aluminosilicate precursor metakaolin with sodium hydroxide (NaOH) and sodium silicate (Na₂SiO₃) as well as the foaming agent was studied for copper ions adsorption from aqueous solution. This paper investigated the effect of adding hydrogen peroxide (H₂O₂) and aluminium powder as foaming agents to an alkali activated materials slurry. The experimental range included 0.50 wt%, 0.75 wt%, and 1.00 wt% hydrogen peroxide and 0.02 wt%, 0.04 wt%, and 0.06 wt% aluminium powder. A control sample without a foaming agent was also created for comparison. The specific surface area, water absorption, density, compressive strength and microstructure of metakaolin based alkali activated materials were evaluated. The adsorption capability of Cu²⁺ with addition of hydrogen peroxide and aluminium powder was then tested. Results indicate hydrogen peroxide addition had superior pore size distribution and homogeneous porosity than aluminium powder, implying improved copper ion elimination. Cu²⁺ adsorption capability reached 98% with 0.75 wt% hydrogen peroxide and 24.6076 m²/g surface area. The results demonstrating that low cost metakaolin-based AAMs are the most effective adsorbent for removing copper ions.

Keywords: Metakaolin; alkali activated materials; hydrogen peroxide; aluminium powder and copper ions adsorption

1. Introduction

Environmental degradation has emerged as one of the most critical issues we face today due to fast and uncontrolled expansion across the globe. Given that water is considered a vital component of life, one of the most important environmental concerns that has attracted worldwide attention is the pollution of water sources such as lakes, rivers, and the sea. Home sewage, industrial waste, and storm water runoff are the most common sources of water pollution [1,2]. In the presence of heavy metals, copper (Cu), lead (Pb), zinc (Zn), iron (Fe), cadmium (Cd), and other ions pollutants are frequently discovered [3-6].

Copper metal is one of the most toxic heavy metals for both humans and the environment [7,8]. Copper is produced in a variety of industries, including metal cleaning and plating baths, paper and paperboard mills, batteries, wood pulp production, tyre manufacture, and fertiliser making [9,10]. Excessive copper heavy metal accumulation in the body may cause major health issues such as organ failure, chronic anaemia, stomach and intestinal inflammation, and liver damage [11-13].

Ion exchange, filtration, chemical precipitation, reverse osmosis, electrochemical treatment, evaporative recovery, and adsorption are all effective methods of copper removal from wastewater. Apart from that, adsorption is recognised as one of the most effective and economical ways of removing heavy metal ions from wastewater owing to its low cost, wide adaptability, easy processing, and excellent efficiency [14,15]. Active carbon [16], clay minerals [17], biomaterials [18], zeolites [19], and industrial solid wastes are some of the materials utilised as adsorbents for metal ions to clean wastewaters using the adsorption approach.

One method for treating wastewater by heavy metal adsorption in water is to use porous metakaolin-based alkali-activated materials, AAMs as an adsorbent. Because of its performance, metakaolin has been widely studied and used as a raw material in the removal of heavy metal ions such as copper, nickel, and zinc ions from aqueous solutions [20,21]. The creation of self-hardening alkali activated materials adsorbents that just need a low curing temperature would make the process more efficient [22]. Amorphous alkali aluminasilicates derived from alkali

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activation or geopolymerization by combining metakaolin with alkaline activators solution [23-28].

Foaming agents, such as hydrogen peroxide or aluminium powder, are frequently added to the AAMs slurry to create gas bubbles and build porous alkali activated materials with a higher surface area, which are important physicochemical properties for determining adsorbent effectiveness in wastewater treatment [29-31]. The contact area between pollutants and adsorbents, which is a vital component in contaminant adsorption, is greatly influenced by pore structure [7]. These porous open-cell alkali activated materials have dramatically improved porosity and permeability, which assists in the transfer and adsorption of contaminants.

Consequently, there is increasing interest in the various pore distribution configurations of alkali activated materials that might be used as heavy metal ion adsorbents. As a consequence, the purpose of this study is to explore further into the potential of metakaolin-based alkali activated materials as a Cu^{2+} adsorbent in aqueous solution at low synthesis temperatures in both types of foaming agents. The addition of a foaming agent to alkali activated materials is meant to assist researchers in better understanding the influence of various types of foaming agents on the formation of porous geopolymers and the percentage removal of Cu^{2+} ions.

2. Experimental method

2.1. Materials

Metakaolin was used as the raw material, while sodium silicate and sodium hydroxide solutions were used as activators. Kaolin that was acquired from Kaolin Sdn. Bhd. Tapah, Perak, Malaysia used in this research as an aluminosilicate material that was calcined at 850°C for 2 hours at $5^\circ\text{C}/\text{min}$ heating rate to generate metakaolin. This calcination temperature was selected based on the optimum calcination profile obtained by [32,33]. Using an X-ray fluorescence spectrometer, the chemical composition of metakaolin was determined. The results are presented in TABLE 1. The total SiO_2 and Al_2O_3 composition of metakaolin is 92.44 percent. The activating solutions sodium hydroxide arrangement (NaOH) with an 8 molar concentration were made by dissolving 320 g of >99% purity NaOH pallets in 1000 ml of pure water. While sodium silicate (Na_2SiO_3) was used as a soluble base activator for alkali activation. To enhance the production of foam, hydrogen peroxide and silver-shaded aluminium powder were used, which is appropriate for the synthesis of porous structure of metakaolin-based AAMs adsorbent. Tween 80 is used as a pore stabiliser to minimise surface tension and drainage in alkali activated metakaolin structures. Copper nitrate, $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ was dissolved in distilled water to evaluate the efficiency of the alkali activated metakaolin adsorbent.

2.2. Preparation

The alkaline activator solution was formed by mixing 8 M NaOH solution with sodium silicate material at a mass ratio of 0.5 $\text{Na}_2\text{SiO}_3/\text{NaOH}$ for at least 5 minutes with a magnetic stirrer. Metakaolin powder was then added to the solution at a constant 0.8 solid to liquid ratio and stirred until homogeneity was attained. The combination was treated with various amounts of hydrogen peroxide (0.50 wt%, 0.75 wt%, and 1.00 wt%) and aluminium powder (0.02 wt%, 0.04 wt%, and 0.06 wt%). The solution was then gently supplemented with 3 wt% of Tween 80 by mass of solid as a surfactant. For comparison, a control sample was generated without the application of a foaming agent. The metakaolin-based alkali activated materials were created in a 1-2 cm spherical shape. 50 mm \times 50 mm \times 50 mm cubic sample are also prepared for compressive strength investigation. The samples were cured at 60°C for 24 hours. After that, the sample was aged at room temperature for seven days before being examined. For the adsorption test, the materials were gently crushed in a pestle and mortar and sieved to achieve a particle size of 150 μm . Analytical grade standard $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ was dissolved in deionised water to make the Cu (II) ion stock solution. It was then diluted to the appropriate concentrations for the experiment. After that, amount of $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ ion stock were diluted in 1000 ml of deionised water in volumetric flask.

2.3. Test and analysis method

The chemical composition of raw material was determined by using PANalytic PW4030 X-ray fluorescence (XRF) spectrometer. The water absorption was measured using Eq. (1) with a standard research procedure in accordance with ASTM C 642-97. The bulk density of the samples was determined using an electronic densitometer MD-3005 equipment in accordance with ASTM D792. The compressive strength of the unfoamed and foamed metakaolin-based AAMs was evaluated following ASTM C109 using Universal Testing Machine (UTM), machine. Three samples were analysed for each parameter.

Next, the specific surface area and pore structure were determined by the Brunauer, Emmett and Teller (BET) methods using Micrometrics Tristar II 3020 volumetric adsorption/desorption apparatus. Following ASTM D3663-20, the amount of nitrogen gas adsorbed by the sample at different low-pressure conditions has been used to measure the surface area of metakaolin AAMs. The microstructural variations of metakaolin, unfoamed alkali activated metakaolin, and foamed alkali activated metakaolin were analysed using a TESCAN TM 3000 with secondary electron detectors. The cut portion of alkali activated metakaolin was used for microstructural investigation after compressive strength testing.

In addition, the phase investigation which is one of the testing procedures used to characterise crystalline materials was carried out using a Shimadzu X-ray diffractometer. For the XRD

study, Cu K radiation scanning from 2θ values in the range of 10° to 80° at a scan rate of 2° per minute and scan increments of 0.02° (2°) was utilised. The XRD pattern was examined using the X'pert HighScore Plus programme and the ICDD PDF-2 database.

$$\begin{aligned} \text{Water absorption (\%)} &= \\ &= \frac{\text{Saturated weight, } W_s - \text{Dry weight, } W_d}{\text{Dry weight, } W_d} \times 100\% \quad (1) \end{aligned}$$

In the adsorption tests, variable metakaolin-based AAMs samples with varying foaming agent (aluminium powder and hydrogen peroxide) concentrations were employed. 0.15 g of sieved metakaolin-based AAMs with different foaming agent concentrations were poured to the conical flask. At room temperature, adsorption experiments for copper ion solution at a concentration of 100 mg/L fix at pH 5 were performed. Small quantities of 0.01 M hydrochloric acid, HCl, and 0.01 M sodium hydroxide, NaOH, were added to keep the pH of the metal ion solutions stable. An orbital shaker was used to shake the flasks for 1 hour at 150 rpm. This batch adsorption condition is based on previous research findings that demonstrated metakaolin-based alkali activated materials or metakaolin geopolymer is an effective adsorbent with favourable Cu^{2+} ion adsorption [20,34,35]. The amount of Cu^{2+} in the samples was then determined using atomic absorption spectroscopy, or AAS. To get the mean findings, the experiment was performed three times for each condition.

3. Results and discussion

3.1. Chemical composition analysis

From TABLE 1, the chemical composition of metakaolin from XRF investigation was shown that metakaolin contains highest measure of silicon dioxide (SiO_2) which was 56.84% followed by aluminium oxide (Al_2O_3) at 35.60%. According to the results, metakaolin satisfied the requirements for usage as a raw material precursor in the development of alkali activation materials. This is because the materials used to manufacture AAMs should be rich in Si and Al, since these elements are sub-

stantial sources of Si^{4+} and Al^{3+} in the binding system that will be activated by an alkali activator solution [36,37]. Reactivity as a source material for geopolymerization increases when more SiO_2 and Al_2O_3 are produced [38]. A tiny amount of magnesium oxide (MgO), sodium oxide (Na_2O), and iron oxide (Fe_2O_3) is found in metakaolin. There was also a trace of potassium oxide (K_2O), titanium dioxide (TiO_2), and calcium oxide (CaO) despite the fact that it was less than 1%.

3.2. Water absorption

The water absorption of metakaolin-based alkali activated materials with varying concentrations of foaming agent is shown in Fig. 1. Water absorption increased from 23.09%, 28.36% and 34.12% when 0.5 wt%, 0.75 wt%, and 1.00 wt% hydrogen peroxide was added, respectively, and from 13.23%, 21.7% and 35.29% at 0.02 wt%, 0.04 wt%, and 0.06 wt% aluminium powder was added. The sample with the highest water absorption (35.29%) had 0.06 wt% aluminium powder by mass of metakaolin, whereas the sample with the lowest water absorption (5.02%) contained 0.00 percent foaming agent. These data clearly show that increasing the amount of foaming agent enhances pore size, which leads to an increase in water absorption value [38]. The addition of foaming agents to alkali activated materials slurry results in a final product with more connected pores that aid in water movement [39,40]. There is an increase in water absorption when the volume of foam and the number of pores that are beneficial to water flow. Researchers have also shown that water absorption percentage increases when foam volume is raised, which supports this conclusion [41,42].

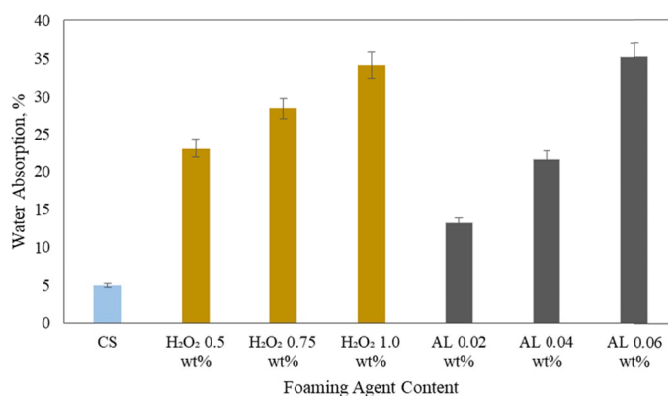


Fig. 1. The water absorption value for metakaolin based alkali activated materials with different foaming agent content

TABLE 1

Percentage of chemical compositions of metakaolin by XRF

Chemical	Mass ratio [%]
SiO_2	56.84
Al_2O_3	35.60
Na_2O	2.40
K_2O	1.04
Fe_2O_3	1.31
TiO_2	0.78
MgO	1.79
CaO	0.24

Another observation was that increasing the quantity of foaming agent in alkali activated materials paste caused an increase in water absorption values [43]. This was due, in part, to the presence of air in the cavities of the large pores, which prevented water from completely filling them. Water permeability increased as the structure became more porous, which might be attributed to the capillary action and the large open spaces between air holes [44-46]. Hence, permeability reduces

as porosity and pore disconnection diminish. In this case, the capacity of a porous material to enable fluids to pass through it is critical for an effective adsorbent.

3.3. Density analysis

Fig. 2 shows the density difference between alkali activated metakaolin samples at various foaming agent percentages (aluminium powder and hydrogen peroxide). In general, when the porosity of the sample increases, the density of the sample decreases. This is due to the fact that aluminium powder and hydrogen peroxide helps to increase the porosity of the sample. According to Fig. 2, the AAMs sample with 0.06% aluminium powder had the lowest density (1.318 g/cm^3) when compared to the others. On the other hand, the metakaolin based AAMs sample with 0.02 wt% aluminium powder had the highest density (1.694 g/cm^3) after the control sample (1.821 g/cm^3). As the quantity of foaming agent in the combination was increased, the density of the mixture reduced, but the porosity increased. This was acknowledged and agreed upon by a significant number of researchers [46,47]. Zhang et al., also stated that by increasing foaming agent concentrations, the porosity of the samples can be shown to rise [48]. This is because the foaming agents released oxygen gas or alumina into the paste, which caused it to break down. This caused more voids to form, which made the alkali-activated materials less dense.

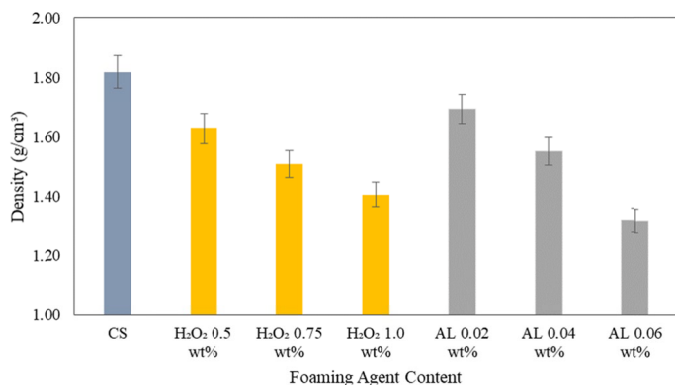


Fig. 2. The density of metakaolin based alkali activated materials with different foaming agent content

3.4. Compressive strength analysis

The average value of the compressive strength evaluated after 7 days of curing is shown in Fig. 3. According to the figure, the control sample had the maximum compressive strength (25.231 MPa) when compared to the other samples that had been foamed. The highest percentage of hydrogen peroxide and aluminium powder added into metakaolin based alkali activated materials produced the lowest compressive strength, which was 13.276 MPa and 8.355 MPa, respectively. An increase in the proportion of foaming agent in metakaolin geopolymer causes a drop in compressive strength. Besides, changes in sample density

corresponded to changes in strength, with lower density samples demonstrating lower strength. It is associated with increases in the quantity of foaming agent used, which leads to an increase in the number of air bubbles in metakaolin geopolymer and pore size in the finished geopolymer resulting in decreased sample strength [49,50].

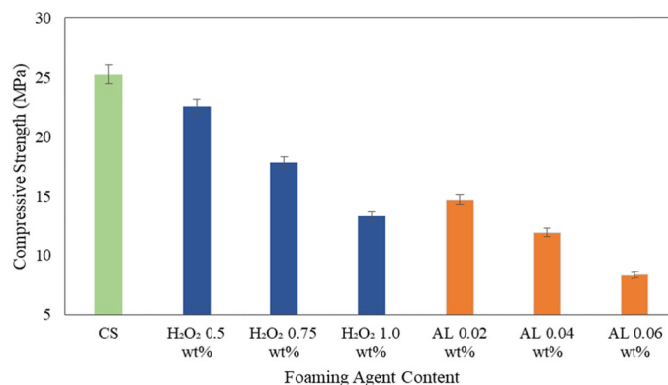


Fig. 3. The compressive strength of metakaolin based alkali activated materials with different foaming agent content

3.5. Pore structure analysis

TABLE 2 summarises the findings of BET study including the specific surface areas (BET), average pore widths, and total pore volumes for metakaolin-based alkali activated materials adsorbents with varying percentage of different foaming agents added. Overall, the addition of a foaming agent enhanced the porosity and the surface area of the alkali activated metakaolin adsorbent. When hydrogen peroxide and aluminium powder is added to a metakaolin-based alkali activated materials, the result is a specimen with a reduced bulk density and a larger apparent porosity [51,52].

Metakaolin based alkali activated materials without addition of foaming agent exhibit $7.1216 \text{ m}^2/\text{g}$ surface area which larger than metakaolin raw powder which is only $4.5752 \text{ m}^2/\text{g}$. Geopolymerization leads to an increase in both the total pore volume and the specific surface area of the material [53,54]. With addition of H_2O_2 at 0.5 percent by mass of metakaolin, the surface area was $10.8483 \text{ m}^2/\text{g}$ and the pore volume was $0.100423 \text{ cm}^3/\text{g}$. The surface area of metakaolin-based AAMs grew to $28.6057 \text{ m}^2/\text{g}$ at 1.0 percent hydrogen peroxide, with the pore volume and average pore width rose to $0.171588 \text{ cm}^3/\text{g}$ and 23.45917 nm , respectively. This metakaolin-based alkali activated materials adsorbent had a specific surface area that was comparable to those reported by Luukkonen et al., using the same metakaolin geopolymer developed for NH_4^+ removal [55]. This was most likely due to the addition of foaming agent to the metakaolin-based alkali activated materials adsorbent that was employed in this study. This enhanced the characterization of the metakaolin-based AAMs adsorbent.

Furthermore, when aluminium powder was added at 0.02 weight percent and 0.06 weight percent rates, the specific surface area rose from $11.2678 \text{ m}^2/\text{g}$ to $19.254 \text{ m}^2/\text{g}$. However, total

TABLE 2

The pore structure of raw metakaolin and metakaolin based alkali activated materials with different percentage of foaming agent added

Sample	Specific Surface Area, m ² /g	Total Pore Volume, cm ³ /g	Average Pore Width, nm
Raw MK	4.5752	0.012366	6.9267
CS	7.1216	0.028246	8.3636
H ₂ O ₂ 0.50 wt%	10.8483	0.100423	20.1631
H ₂ O ₂ 0.75 wt%	24.6076	0.171588	18.54188
H ₂ O ₂ 1.00 wt%	28.6057	0.194348	23.45917
AL 0.02 wt%	11.2678	0.116334	9.24562
AL 0.04 wt%	17.1592	0.085391	15.38298
AL 0.06 wt%	19.254	0.071427	29.34117

pore volume decreased as pores expanded in size and interconnected, resulting in pore coalescences. When the foaming agent concentration rose, the specific surface area and average pore width distribution of alkali activated materials improved in line with [56,57]. Tan et al., stated that the porosity of the alkali activated material adsorbents enhanced adsorption efficiency and rate [58].

3.6. Microstructural analysis

The amount of hydrogen peroxide and aluminium powder present in a metakaolin-based alkali activated materials influences its microstructure illustrated in Fig. 4. The control sample

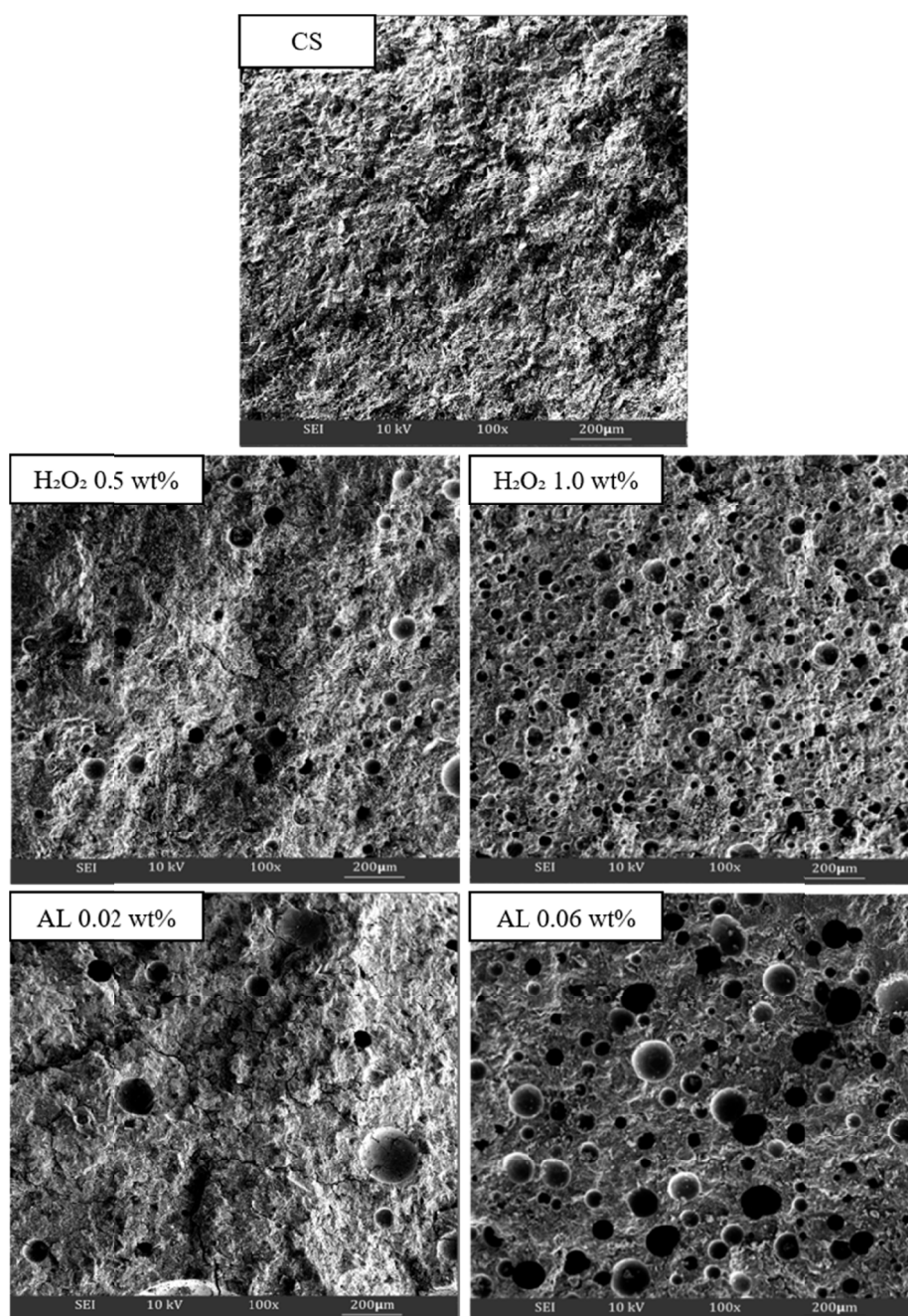


Fig. 4. Microstructural of metakaolin based alkali activated materials at different percentages of foaming agent

had the densest AAMs matrix of all the samples since no foaming agent was applied that presented only tiny pores. While gaps and porosities were readily visible in alkali activated metakaolin samples treated with a foaming agent (aluminium powder and hydrogen peroxide). Basically, aluminium powder reacted with water in an alkaline solution to produce hydrogen gas (H_2) and aluminium hydroxide ($Al(OH)_3$) whereas hydrogen peroxide decomposes gradually into water and oxygen gas, causing gaps and holes to form.

With the addition of a foaming agent, the metakaolin-based AAMs formed a loose and porous structure that diffused equally throughout the matrix. H_2O_2 and Al powder were added to increase porosity with wider average pore diameter. As the concentration of H_2O_2 grew, the geometric shape of the pores changes [59,60]. All pores were spherical in form, homogenous in structure at low H_2O_2 concentrations (0.5 wt%). The narrow pore size range, as previously stated, was responsible for the increased compressive strength. The presence of high H_2O_2 and Al concentrations boosted the probability of generating larger holes or macropores. The 0.06 wt% sample has a greater proportion of big pores due to the high level of foaming in these samples, the pore walls were very thin, allowing for the fusing of some pores and the development of these massive voids.

3.7. Phase analysis

The XRD diffractogram of raw metakaolin and metakaolin based alkali activated materials (Fig. 5) revealed typical broad

amorphous hump around 20° - 40° 2θ . Quartz was identified as the element that caused the strong distinctive peak that was discovered in metakaolin-based alkali activated materials. In terms of the X-ray diffractogram of metakaolin, the quartz is caused by metakaolin, however this quartz does not participate in the chemical process itself. The primary phases of MK are muscovite ($KAl_2(Si_3Al)O_{10}(OH,F)_2$), quartz (SiO_2), and kaolinite ($Al_2Si_2O_5(OH)_4$). The presence of silica and alumina is indicated by the presence of quartz and muscovite in the structure [61]. As it activated during the alkali activation process, the kaolinite peak in MK reduced in metakaolin-based alkali activated materials. The illite phase at 35° persisted in the crystalline structure in the control sample, comparable to the results of [62]. The illite structure coexisted with metakaolin, which when a geopolymer is formed, the surface becomes denser, indicating a build-up of illite or mullite phase. As a consequence, increasing the amount of hydrogen peroxide in metakaolin-based AAMs did not produce significantly different formation peaks in the XRD diffractogram. This suggests that the quantity of H_2O_2 applied to the AAMs has no influence on the alkali activation or geopolymerization process. However, as claimed by Ariffin et al., the addition of aluminium powder lowers the amount of alkali activator solution in the AAMs [63], enabling the AAMs adsorbent to have a more crystalline phase structure, as observed scattered diffraction peaks appearing at 18 - 35° 2θ in AL 0.06 wt% sample. In line with Zhang et al., the crystallisation of a material will result in an increase in the viscosity, consequently restricting the material expansion and resulting in a greater surface tension that affects the development and stability of pores [64]. On the other hand,

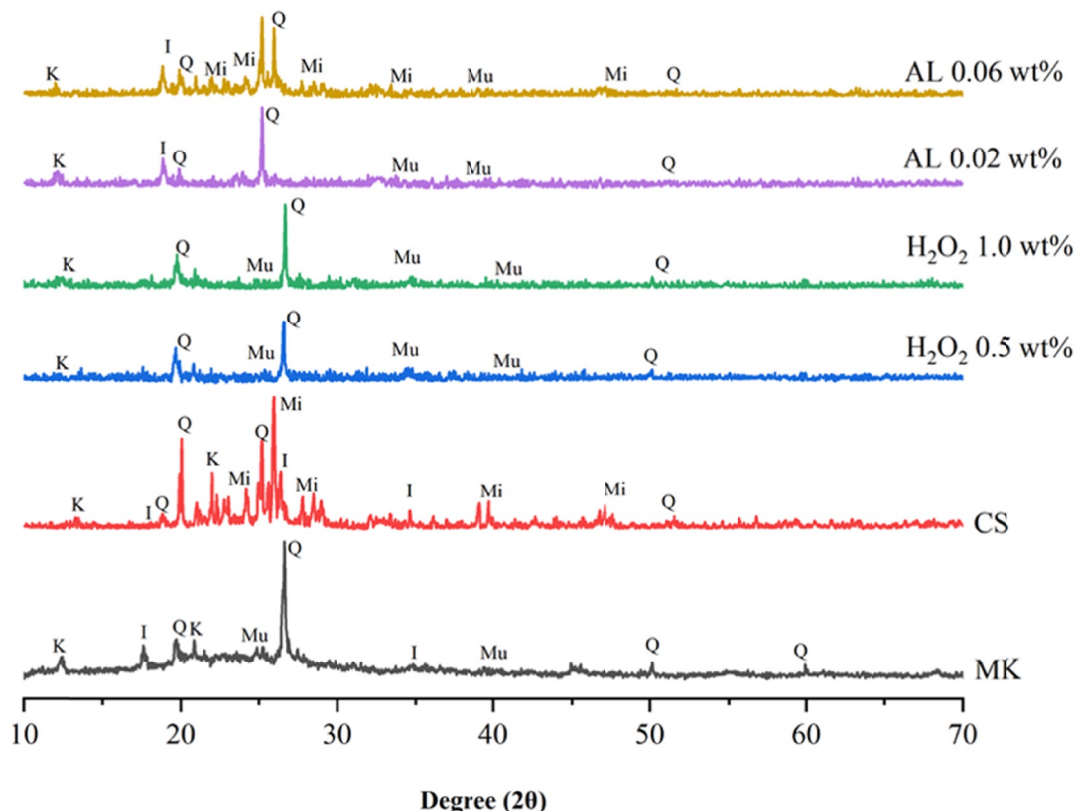


Fig. 5. Phase analysis of metakaolin based alkali activated materials at different percentages of foaming agent

muscovite phase was found in metakaolin-based AAMs when 0.5, 1.0 wt% of hydrogen peroxide and 0.02 wt% aluminium powder were added. Due to micas have distinctive characteristics, muscovite, a micaceous mineral, is one of the natural minerals that may block radioactive heavy metals in the environment, and their adsorption capacity method are very divergent [65,66]. According to Wu et al., muscovite has a higher rate of Sr(II) adsorption at lower pH values than biotite [67]. Furthermore, these samples (H₂O₂ 1.00 wt% and AL 0.02 wt%) approach to zeolites in which they are amorphous or semi-crystalline and have heterogeneous interior micro-pores, making them favourable for use as an adsorbent [68].

3.8. Effect on foaming agent on copper ions adsorption

To improve the porosity of alkali activated metakaolin and copper adsorption by metakaolin based AAMs adsorbents, hydrogen peroxide and aluminium powder that created hydrogen gas were used as foaming agents. As shown in Fig. 6, the effect of adding a foaming agent to metakaolin-based AAMs adsorbent on copper ion adsorption by the metakaolin-based AAMs adsorbent was examined. The control sample had the lowest copper removal efficiency, at 20.3%. This is because the control sample lacks a porous structure when compared to the sample treated with hydrogen peroxide and aluminium powder. With a removal rate of 98.14%, the greatest removal was achieved at a concentration of 1.00 wt% hydrogen peroxide. The increased Cu²⁺ adsorption indicates that the alkali activated metakaolin adsorbent efficiency has improved with the increasing of hydrogen peroxide percentage. A considerable percentage was also obtained at 0.06 wt% Al powder. On the other hand, consumption did not achieve the maximum percent removal. However, when aluminium powder doses rose by 0.08 wt%, the removal efficiency went down, and the removal amount dropped to 70.3%. This attributed to the excessive quantity of aluminium coupled with silica and the coordination number of aluminium in the materials also had an effect on how the materials bonded together at the end [63,69].

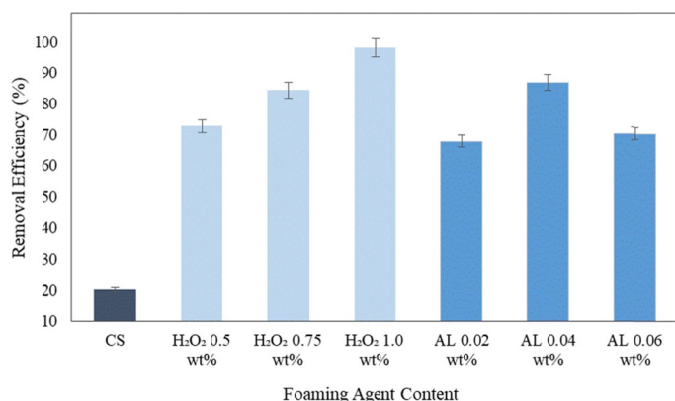


Fig. 6. Percentage removal of Cu²⁺ at different percentages of hydrogen peroxide and aluminium powder addition

4. Conclusion

The effects of hydrogen peroxide and aluminium powder on the adsorption of copper ions by metakaolin-based alkali activated materials adsorbent have been investigated. The adsorption potential and porosity of metakaolin-based AAMs adsorbent structures varied depending on the proportion of foaming agents used. A simple method was applied to produce the metakaolin-based AAMs adsorbent at 60°C. This is lower than the sintering temperature of most zeolite or heavy metal removal membranes. Since the metakaolin based AAMs adsorbent has a well-defined pore size distribution and a lot of total porosity, it has a lot of surface area that can be used to adsorb copper ions. Adding a foaming agent to the alkali activated metakaolin adsorbent is meant to make the adsorbent more beneficial for removing copper ions from water than the control sample without the foaming agent. Through the adsorption method, 0.15 g of metakaolin-based AAMs adsorbent in 1.0 wt% hydrogen peroxide removed the most Cu²⁺ (98.14%). The adsorption technique utilizing hydrogen peroxide foaming agent was shown to be the most successful in adsorbing copper ions when compared to aluminium powder because hydrogen peroxide formed more homogeneous and controlled pores, which improved the attributes of excellent adsorbents.

Acknowledgement

The author would like to acknowledge the support from the Fundamental Research Grant Scheme (FRGS) under a grant number of FRGS/1/2019/TK10/UNIMAP/02/21 from the Ministry of Education Malaysia.

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