



## RESEARCH ARTICLE

### An investigation on the application of CaII-FeII-FeIII magnetic hydrotalcite-like-compounds for the removal of impurities in aqueous systems

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## ABSTRACT

The main aim of this study is to investigate the feasibility of utilizing novel hydrotalcite-like-compounds as ion-exchangers for the removal of anionic impurities from aqueous solution. Hydrotalcite is naturally occurring, rarely found anionic clay. It has a positively charged layered structure consisting of two metal oxide layers and an interlayer of carbonate anions. Hydrotalcites can, therefore, be used as ion-exchangers. However, the carbonate anion is highly selected by natural hydrotalcite, making the ion-exchange capacity for other anions very low. In this study, several synthetic hydrotalcite-like compounds and a magnetic hydrotalcite-like compound samples were prepared in order to remove anionic impurities from waters. The physical structure and chemical properties of these anionic clays were characterized using standard characterisation techniques. The removal capacities of the synthetic products obtained were then investigated. Magnetic hydrotalcite-like-compound, namely, CaII-FeII-FeIII, was proved to be a good ion-exchanger.

**Keywords:** Double hydroxides, clay characterization, anionic impurities

## 1. INTRODUCTION

Hydrotalcite is naturally occurring, rarely found anionic clay. It has a positively charged layered structure consisting of two metal oxide layers and an interlayer of carbonate anions. The structure of hydrotalcite has been investigated by a number of researchers. Allmann [1] and Taylor [2] reported the structure of hydrotalcite by as  $Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$ . Hydrotalcite has a layered structure consisting of  $CO_3^{2-}$  anions sandwiched by two metal layers [3]. The structure of hydrotalcite is very similar to the naturally occurring form of magnesium hydroxide ( $Mg(OH)_2$ ) known as brucite. Because of its layered form, it is also called a 'Layered Double Hydroxide' (LDH). Natural hydrotalcite ( $Mg^{2+}$  and  $Al^{3+}$  as cations) is a member of LDH group of minerals [4, 5], which can be typified by the minerals pyroaurite and sjögrenite ( $Mg^{2+}$  and  $Fe^{3+}$  as cations) [6-8].

Normally, hydrotalcite stays neutral, however, randomly divalent ions ( $Mg^{2+}$ ) are substituted by

trivalent ions ( $Al^{3+}$ ) generating positively charged layers [9]. These layers can be called host layers. This positive charge is compensated by  $CO_3^{2-}$  anions and water molecules between the two metal layers. The interlayer can be named as a guest layer because of its ion-exchange properties. Water molecules in the interlayer have hydrogen bonds with both carbonate anions and metal sheets [10].

Synthetic hydrotalcite-like compounds have been widely investigated since 1970s. Recently, there is a growing interest for the use of hydrotalcite-like compounds in the removal of aquatic inorganic and aromatic pollutants [11-13]. The general formula of hydrotalcite-like compounds is given as  $[M^{2+}M^{3+}(OH)_2]^+ \times [A^{n-} \cdot mH_2O]$  where A represents an exchangeable anion such as  $CO_3^{2-}$ ,  $SO_4^{2-}$ ,  $NO_3^-$  or  $Cl^-$ , having a valence of n [14, 15]. The divalent metal ion could be  $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Zn^{2+}$  or  $Ni^{2+}$ , and the trivalent metal ion  $Al^{3+}$ ,  $Fe^{3+}$  or  $Cr^{3+}$ .

The ratio of divalent metal ion to trivalent metal ion differs according to the mineral, but, in general,

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minerals belong to the pyroaurite-sjögrenite group have a ratio of 3:1 [6]. When made synthetically, M(II)/M(III) ratio could take values between 1.0 to 4.0 [16, 17].

Due to the positively charged structure of hydrotalcites, the interlayer carbonate anion can be exchanged with other anions. Hydrotalcites can, therefore, be used as ion-exchangers. However, the carbonate anion is highly selected by natural hydrotalcite, and the access to the interlayer region by other anions is restricted [18]. Hydrotalcite-like compounds that are prepared carefully under a  $\text{CO}_3^{2-}$ -free environment can show significant anion-exchange capacities [19]. Jones and Chibwe [20] and Carlino [21] have reported anion-exchange capacities up to 4.1 meq  $\text{g}^{-1}$ . Therefore, compared to natural cation exchangers with cation-exchange capacities of 0.7 to 1.0 meq  $\text{g}^{-1}$  [21, 22], hydrotalcite-like compounds generally have a higher ion-exchange capacity. Numerous hydrotalcite-like-compounds have been synthesized so far in order to remove inorganic [13, 23, 24] or organic impurities [25-28] in aqueous solutions.

The magnetic CaII-FeII-FeIII type hydrotalcite-like clay mineral was synthesized in this study, in order to investigate the feasibility of utilising these derived products as an ion-exchanger, and for the easy removal of this product from the batch tank, as these clay minerals have too small particle sizes to settle with their own velocity.

## 2. MATERIALS AND METHODS

### 2.1. Synthesis of hydrotalcite-like compounds

For the preparation of hydrotalcite-like compounds, the method used by Reichle [29] was employed with slight modifications. The reason to synthesize hydrotalcite-like clays was because of hydrotalcite's selectivity for the carbonate ion. In this study, three different hydrotalcite-like compounds and one magnetic hydrotalcite-like compound have been synthesized. In these compounds, the divalent metal ion was  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Fe}^{2+}$ , the trivalent ion was  $\text{Al}^{3+}$  or  $\text{Fe}^{3+}$  and the interlayer anion was  $\text{Cl}^-$  ion. In this paper, the hydrotalcite-like compounds were named in the form of "A-B-C", where A is the divalent metal ion, B represents the trivalent metal ion and C is the original interlayer anion.

For the synthesis of Mg-Al-Cl, a mixture of 3 M  $\text{MgCl}_2 \times 6\text{H}_2\text{O}$  and 1 M  $\text{AlCl}_3 \times 6\text{H}_2\text{O}$  was prepared outside the reactor. This solution was added dropwise to 4 M NaOH while the mixture was stirred vigorously using a mechanical stirrer. The temperature was kept around 35 °C during mixing. The mixing lasted around 30 minutes and the pH of the reaction mixture was adjusted to 8.5. The sample was then aged for 18 hours at 65 °C in a water bath. After ageing, the slurry was washed repeatedly using boiled cooled distilled water until it was free of ions. The slurry's initial conductivity was 10  $\mu\text{S}$ , and this was reduced to approximately 0.05  $\mu\text{S}$ . The sample was then dried at 110 °C in an air oven. The white powder-like product

was ground and stored in an air-tight sample container in a desiccator.

The synthesis procedure of Ca-Al-Cl was exactly the same with the synthesis of Mg-Al-Cl, except 3 M of  $\text{CaCl}_2 \times 6\text{H}_2\text{O}$  was used instead of 3 M  $\text{MgCl}_2 \times 6\text{H}_2\text{O}$ . Likewise, the synthesis procedure of Ca-Fe-Cl was exactly the same with Ca-Al-Cl. This time the trivalent metal ion was changed to  $\text{Fe}^{3+}$  using  $\text{FeCl}_3 \times 6\text{H}_2\text{O}$  instead of  $\text{AlCl}_3 \times 6\text{H}_2\text{O}$ .

### 2.2. Synthesis of magnetic hydrotalcite-like compound

As the name implies, synthetic magnetite ( $\text{Fe}_3\text{O}_4$ ) or magnetic floc which has a standard formula of  $\text{AB}_2\text{O}_4$  has been used in the water treatment processes as a coagulant [30]. The letters A and B represents ferrous ( $\text{Fe}^{2+}$ ) and ferric ( $\text{Fe}^{3+}$ ) ions, respectively. The advantage of magnetic floc over the common coagulants (e.g.  $\text{FeCl}_3$ ,  $\text{FeSO}_4$ , etc.) is that it can be recovered easily from solution by means of a magnetic separator.

In order to obtain a hydrotalcite-like compound with magnetic properties, equal amounts of ferric and ferrous salts were mixed and the pH was adjusted to 11.0 using NaOH. 3 M of  $\text{CaCl}_2 \times 6\text{H}_2\text{O}$ , 0.5 M  $\text{FeCl}_2 \times 4\text{H}_2\text{O}$  and 0.5 M  $\text{FeCl}_3 \times 6\text{H}_2\text{O}$  were mixed and added into 4 M NaOH solution. The rest of the synthesis procedure was kept the same as Ca-Al-Cl. In order to measure magnetism, the magnetic attraction factor was measured and found as 0.95 which was very close to 1.

### 2.3. Characterisation methods

#### Zeta Potential Analysis

In order to determine the sign charges of the clay mineral particles, Zetasizer 4 (Malvern Instruments) was used. To measure the zeta potential values of the clay samples, 0.25 g of clay sample was suspended in 250 mL 0.01 M  $\text{KNO}_3$  solution. This suspension was then homogenized in an ultrasonic bath for 20 minutes. The pH adjustments were done using KOH and  $\text{HNO}_3$  solution at different concentrations. The readings were recorded at pH values from 2 to 12.

#### X-Ray Diffraction Analysis, XRD

X-ray diffraction is used to obtain information about the structure and the degree of crystallinity of clay samples. The samples were scanned by a Philips PW1830 diffractometer system using Cu K radiation. The diffractometer was fitted with a PW1820 goniometer, an automatic divergence slit and a graphite monochromator. All the samples were scanned over the angular range  $1.6^\circ - 40^\circ 2\theta$  at  $0.15^\circ$  per second.

#### Nitrogen Adsorption Analysis, BET

This is a standard procedure for the determination of the surface area of finely divided and highly porous materials. By determining the nitrogen adsorption

and desorption isotherm of the porous materials, information such as their BET surface area, pore size and microporosity can be obtained. Approximately 0.1- 0.2 g of the samples, previously dried in oven at 110 °C, were degassed at 150 °C at a constant pressure for 3-4 hours and the isotherms were recorded at 77 K. The nitrogen adsorption and desorption isotherms of the samples were recorded on an ASAP 2000 Micromeritics apparatus.

### Particle Size Analysis

The particle sizes of hydrotalcite-like compounds and their derivatives were determined using a Malvern Instruments-1200 Particle Sizer and Analyser.

### 2.4. Data treatment

A plot of concentration on the adsorbent against equilibrium concentration in the solution at constant temperature is referred to an isotherm. A number of isotherms are available to describe adsorption behaviour. However, there are three common isotherms used for adsorption, which are the Langmuir, Freundlich and BET isotherms. The isotherms for adsorption from aqueous media generally follow a monolayer adsorption isotherm, although under some conditions multilayer adsorption has been encountered [31]. The monolayer adsorption isotherms have also been used to describe the ion-exchange behaviour of anions. The Langmuir model has been used for phosphate [32], chromate [33] and heavy metal [34] removal using ion-exchangers. The Langmuir isotherm can therefore be defined as one of the classic adsorption isotherms used for ion-exchange correlations. The Langmuir isotherm assumes two hypotheses which are (1) adsorption is on a monolayer (i.e. there is a fixed number of accessible sites on the adsorbent) and (2) adsorption is reversible. Only when the rate of adsorption of molecules on to the surface is the same as the rate of desorption of molecules from the surface, equilibrium is reached. The Langmuir isotherm corresponds to monolayer adsorption and is expressed as follows (Eq. 1);

$$q = \frac{Q^{\circ} b C_{eq}}{1 + b C_{eq}} \quad (1)$$

In this equation  $q$  is the concentration of the species on the adsorbent ( $\text{mg g}^{-1}$ ),  $C_{eq}$  is the equilibrium concentration of solution ( $\text{mg L}^{-1}$ ) and  $Q^{\circ}$  and  $b$  are the Langmuir constants.  $Q^{\circ}$  represents the saturation concentration of adsorbed species on the adsorbent, i.e. maximum monolayer adsorption capacity. The Langmuir constant  $b$  is related to the enthalpy of adsorption. Calculating this constant is important because it represents the binding strength of the adsorbent. The isotherm equation can be rearranged as seen in Equation 2.

$$\frac{1}{q} = \frac{1}{Q^{\circ}} + \frac{1}{b Q^{\circ}} \times \frac{1}{C_{eq}} \quad (2)$$

Plotting  $1/q$  against  $1/C_{eq}$  should give a straight line with a positive intercept. By using the gradient and the intercept on y-axis the Langmuir constants  $Q^{\circ}$  and  $b$  can be calculated.

### 2.5. Experimental procedure

All batch-technique experiments were carried out in 500 mL conical borosilicate glass flasks. All chemicals were supplied by BDH Chemicals Co. Ltd. in analytical reagent grade and were used without further purification. The adsorbate solutions were prepared with de-ionized water. The stock solutions (1 M) were prepared and stored accordingly. Necessary dilutions at a different range of concentrations ( $10\text{-}300 \text{ mg L}^{-1}$ ) were prepared prior to the experiments and a control sample was also prepared. A known amount of adsorbent, 0.1 (for chromate) or 0.25 g (for permanganate), was weighed carefully and placed in conical flasks containing solution at desired concentration. The sample solutions were then contacted with adsorbent for 18 hours. The ambient temperature was kept at 16-17 °C. The suspensions were centrifuged at 3500 rpm for 20 minutes in order to separate the adsorbent from the slurry. It should be noted that 0.1 g of adsorbent is equivalent to approximately 2000 particles of hydrotalcite, except for samples with magnetic property, where 0.1 g of adsorbent is equivalent to approximately 1500 particles. Hydrotalcite's and magnetite's densities were reported to be 1.929 and 5.197  $\text{g cm}^{-3}$ , respectively [35]. Since a large number of particles were taken for the experiments, there is a low error due to sampling. Perkin-Elmer 1100B AAS (Atomic Absorption Spectrometry) was used to determine chromium and manganese concentrations.

## 3. RESULTS AND DISCUSSION

### 3.1. Characterisation results

The electrophoretic curves of hydrotalcite-like compounds and modified hydrotalcite were given in Fig 1. All samples showed anion exchanger behaviour with an obvious trend. Zeta potential values decrease with an increase in pH, indicating higher anion exchange capacities at lower pH. It should be noted that there is little variation in the zeta potential for the Mg-Al-Cl clay for a range of pH values. Modified hydrotalcite showed very good potential of anion exchange behaviour at low pH, however it apparently loses its anion exchange properties above pH 7.

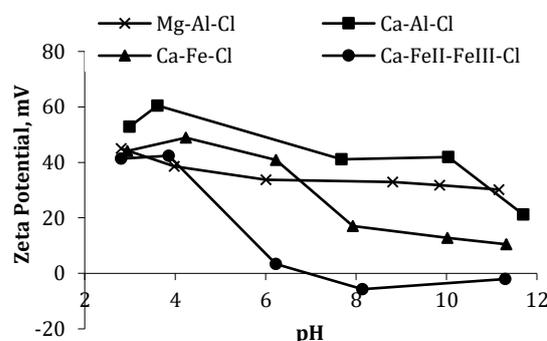


Fig 1. Zeta potential of hydrotalcite-like compounds

A summary of XRD analysis results of hydrotalcite-like compounds can be found in Table 1. The XRD analysis result of synthetic hydrotalcite was also given in this

section for comparison purposes. The spacing of  $d_{(003)}$  plane for synthetic hydrotalcite is 7.65 Å, which is in good agreement with synthetic hydrotalcites prepared by other researchers. The  $d_{(003)}$  spacing of natural hydrotalcite was reported to be 7.67 Å [16]. The lattice parameters ( $a_0$  and  $c_0$ ) of synthetic hydrotalcite can be calculated using the  $d_{(003)}$  and  $d_{(110)}$  reflections. The angular position of the first peak,  $d_{(003)}$ , gives an approximate value for the inter-lamellar distance ( $c_0 = 3 \times d_{(003)}$ ), and the  $d_{(110)}$  peak situated around 60° allows the estimation for the cell dimension,  $a_0$ , which is related to the metal-metal inter-atomic distance within sheet ( $a_0 = 2 \times d_{(110)}$ ) [36].

The X-ray diffractogram of synthetic hydrotalcite (Mg-Al-CO<sub>3</sub>) showed very narrow peaks, indicating a good degree of crystallisation. The (003), (006), (110) and (113) reflections represented a symmetrical pattern, however (003) and (006) peaks were higher and sharper compared to other reflections, which indicated a stacked layer structure. The (102), (105) and (108) reflections were broad and asymmetric which typified hydrotalcite phases. The XRD spectrum of synthetic hydrotalcite was used as a reference spectrum.

The XRD diffractogram of Mg-Al-Cl represented a very similar pattern to synthetic hydrotalcite, however it was noted that  $d_{(003)}$  and  $d_{(006)}$  facings were broader compared to the pattern of synthetic hydrotalcite. When the XRD patterns of Ca-Al-Cl and Ca-Fe-Cl type clay samples were observed, it was seen that there were many peaks. These peaks were investigated using X-ray diffractogram database software. When these peaks related to hydrotalcite, phases were eliminated by the computer programme. It was seen that there were peaks showing CaCl<sub>2</sub>, Ca(OH)<sub>2</sub>, AlCl<sub>3</sub>, Al(OH)<sub>3</sub> and FeCl<sub>3</sub> attached to the surface of the clay samples, resulting a loss of crystallinity. These peaks for Ca(OH)<sub>2</sub> and Al(OH)<sub>3</sub> indicated that co-precipitation might occur on the surface of the clay samples during the removal of anionic impurities from water. It was also noted that Ca-Al-Cl and Ca-Fe-Cl samples were not as crystalline as Mg-Al-CO<sub>3</sub> or

Mg-Al-Cl samples. The XRD diffractogram of magnetic hydrotalcite-like compound did not exhibit hydrotalcite phase, but magnetite (Fe<sub>3</sub>O<sub>4</sub>) phase with a distinctive peak at the (111) position.

Nitrogen adsorption analysis was used to determine BET surface area, pore size and porosity of clay samples. Fig 2 shows the average pore diameter of the clay samples. As can be seen from Fig 2, the hydrotalcite-like compounds have surface areas close to each other, except Mg-Al-Cl. Differences in the surface areas between the hydrotalcite-like compounds is due to the divalent metal ions used during the synthesis. For instance, the Ca<sup>2+</sup> ions have higher ionic radii compared with the Mg<sup>2+</sup> ion resulting in a lower BET surface area. The effective ionic radii ( $r_{eff}$ ) of some ions can be seen in Table 2. The larger sized ions occupy a greater volume of the clay surface leading to lower BET surface areas [37]. It was observed that as surface area decreased, the average pore diameter increased for the samples synthesized. The average pore diameter of modified hydrotalcite-like compounds with a magnetic property was larger compared to other hydrotalcite-like compounds.

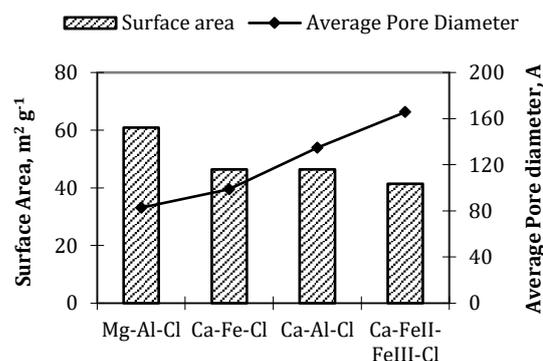


Fig 2. The surface area and average pore diameter of hydrotalcite-like compounds

Table 1. A summary of XRD results for synthetic hydrotalcite and hydrotalcite-like compounds

Compound Name	X-ray Angle (°2θ)	$d_{(003)}$ (Å)	X-ray Angle (°2θ)	$d_{(110)}$ (Å)	$c_0$ (Å)	$a_0$ (Å)
Mg-Al-CO <sub>3</sub>	11.3	7.65	60.6	1.53	23.0	3.06
Mg-Al-Cl	11.3	7.67	60.5	1.51	23.0	3.02
Ca-Al-Cl	11.3	7.81	60.5	1.53	23.4	3.06
Ca-Fe-Cl	11.4	7.79	60.5	1.52	23.4	3.04

$c_0 = 3 \times d_{(003)}$ ;  $a_0 = 2 \times d_{(110)}$

Table 2. Ionic radii of some metal ions and anions [40]

Ion	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Al <sup>3+</sup>	Fe <sup>2+</sup>	Fe <sup>3+</sup>	Cl <sup>-</sup>
Ionic radius, pm	86	114	68	92	79	22

The particle size tests were conducted with a lens which could measure particles with a size less than 188 μm. According to cumulative under size curves, most of the samples were under 3 μm. The results have also shown that most of the particles were in the

range of 1.0-3.0 μm (Table 3). It has been reported that there is an inverse relationship between particle size and surface area [38] and the results confirm this statement. It can be seen that an increase in the particle size resulted in lower specific surface areas.

**Table 3.** The mean diameters and specific surface areas of hydrotalcite-like compounds

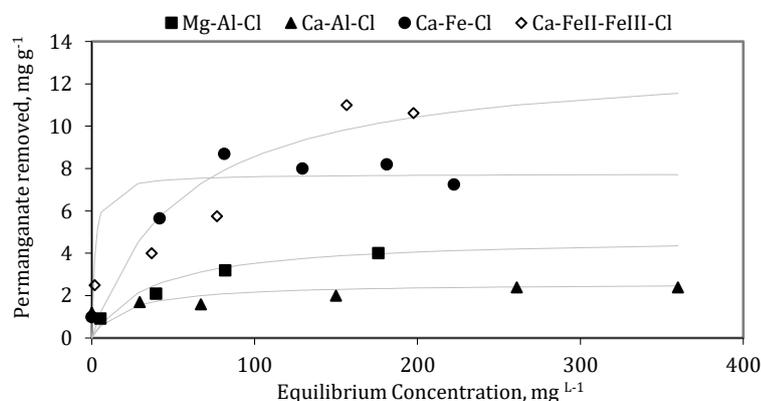
Compound name	Mean diameter, $\mu\text{m}$	Specific Surface Area, $\text{m}^2 \text{cm}^{-3}$
Mg-Al-Cl	1.19	0.36
Ca-Al-Cl	2.50	0.69
Ca-Fe-Cl	2.45	0.42
Ca-FeII-FeIII-Cl	2.70	0.44

**3.2. The ion-exchange experiments**

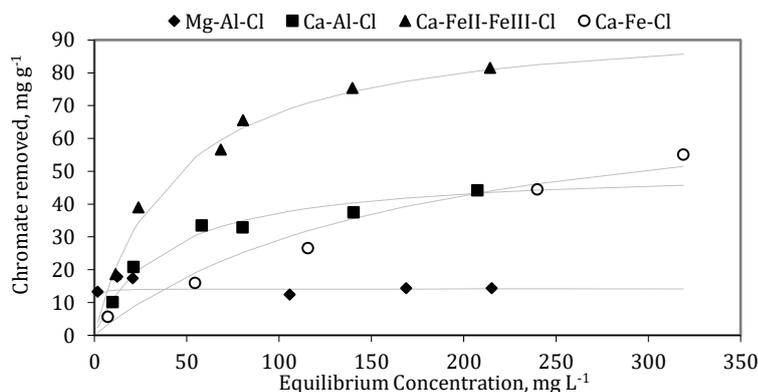
The ion-exchange experiments were carried out using permanganate and chromate solutions. Permanganate can be found in the effluents of several industries. Permanganate ion needs to be removed from waters for aesthetic reasons because it is a strongly coloured ion. The other reason for selecting this ion was that permanganate salts are usually soluble and non-hydrolysable which can be used for ion-exchange capacity determinations. As it is known, strontium is a stable ion used in the determination of the capacity of cationic clays. The permanganate ion was used as its counterpart. The only disadvantage of permanganate ion is that it can be easily oxidized in strong light and is slightly unstable. Unlike potassium permanganate solutions, potassium dichromate solutions are generally stable. However, chromates exist in two different ionic states. These ionic states can occur between pH 0 and 7.3 as dichromate and between pH 7.3 and 14 as chromate [39]. Unfortunately, both

states can be encountered in aquatic systems. During the experiments, the pH of the solution varied from 5.1 to 5.6 and 7.0 to 8.2 for  $\text{K}_2\text{Cr}_2\text{O}_7$  and  $\text{KMnO}_4$ , respectively. The pH measured for  $\text{K}_2\text{Cr}_2\text{O}_7$  solution indicated that dichromate was the dominant ion in the solution. The initial pH of the  $\text{KMnO}_4$  solution increased to above neutral values during the ion-exchange process.

During the experiments, the pH of the solution varied from 5.1 to 5.6 and 7.0 to 8.2 for  $\text{K}_2\text{Cr}_2\text{O}_7$  and  $\text{KMnO}_4$ , respectively. A comparison of adsorption isotherms for hydrotalcite-like compounds can be seen in Fig 3 and Fig 4. The data obtained from the contact tests appear to correlate well with Langmuir model. The transformed data for all the samples gave a straight line with small error,  $R^2 (>0.995)$ . For  $\text{MnO}_4^-$  solution, Ca-Al-Cl showed the lowest capacity compared to other clay samples used. Although effective removal capacities were achieved with Mg-Al-Cl and Ca-Fe-Cl, the highest uptake was attained when the Ca-FeII-FeIII-Cl was used.



**Fig 3.** Adsorption isotherm plots of permanganate for different hydrotalcite-like compounds



**Fig 4.** Adsorption isotherm plots of chromate for different hydrotalcite-like compounds

For  $\text{Cr}_2\text{O}_7^{2-}$  solution, quite similar results were obtained. While Mg-Al-Cl showed the lowest adsorption capacity, Ca-Al-Cl and Ca-Fe-Cl uptake rates were close to each other, with around  $40 \text{ mg g}^{-1}$  capacity. However, as expected, the highest uptake was attained when the Ca-FeII-FeIII-Cl was used. All the results are presented in Table 4. The main reason

for the highest uptake of the analytes on the Ca-FeII-FeIII-Cl was due to co-precipitation on the surface of the clay samples. Although the crystallinity of the magnetic hydrotalcite-like clay was lower compared to the other samples prepared, it showed anion exchange capacities.

**Table 4.** The ion-exchange experimental results

Solution	Ion-exchanger	$Q^\circ, \text{mg g}^{-1}$	$Q^\circ, \text{meq g}^{-1}$	b, $\text{L mg}^{-1}$
$\text{MnO}_4^-$	Mg-Al-Cl	4.70	0.04	0.028
	Ca-Al-Cl	2.51	0.02	0.053
	Ca-Fe-Cl	7.76	0.07	0.550
	Ca-FeII-FeIII-Cl	13.33	0.11	0.018
$\text{Cr}_2\text{O}_7^{2-}$	Mg-Al-Cl	14.14	0.07	0.090
	Ca-Al-Cl	50.25	0.23	0.027
	Ca-Fe-Cl	78.74	0.37	0.006
	Ca-FeII-FeIII-Cl	98.04	0.45	0.023

#### 4. CONCLUSIONS

An investigation on CaII-FeII-FeIII hydrotalcite-like-compound for the removal of impurities in water was carried out in this study. The results of zeta potential analysis gave an indication of anion-exchange properties for the hydrotalcite-like compounds synthesized. The diffractograms obtained from XRD analysis of hydrotalcite samples indicated fairly good crystallinity and all hydrotalcite-like compounds analysed, except the one with magnetic properties, were identified as hydrotalcite structures. However, modified hydrotalcite-like compounds did not exhibit hydrotalcite phases. It has been found that hydrotalcite-like compounds have lower surface areas compared to synthetic hydrotalcite given in literature.

The overall results indicated that hydrotalcite samples prepared are able to remove anionic impurities from water. The above findings demonstrated that magnetic hydrotalcite sample (CaII-FeII-FeIII) have significantly higher capacity compared to the other hydrotalcite-like compounds prepared. Among the hydrotalcite-like compounds, Ca-Fe-Cl is the most cost-effective sample. However, magnetic hydrotalcite, CaII-FeII-FeIII, was also proved to be a good ion-exchanger. Due to easy removal of magnetic hydrotalcite by means of a magnetic separator, this anionic clay would a good material for further study.

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