



Studies on synthesis, characterisation and effect of calcination on Mg-Al hydrotalcite for the removal of anionic species

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Received 23 September 2020; accepted 9 December 2020

The use of Mg-Al Hydrotalcite for removing anionic contaminants such as Bromide, Sulphate, Nitrate, Fluoride and Chloride from aqueous solutions has been studied. The current study analyses the effect of calcination on the anionic removal efficiency of hydrotalcite. The hydrotalcite samples have been calcined batch-wise in a muffle furnace at different temperatures and are added to various anionic solutions in a conical flask and kept for stirring in a shaking incubator for three hours. The filtrate obtained is then analysed using Ion chromatography. The results obtained show a steady increase of anionic removal with an increase in calcination temperature. All anions under study show maximum removal towards calcinated samples than non-calcinated samples. For both calcinated and non-calcinated samples, it is found that the percentage removal of anions decreases with the increase of its ionic radii in the manner $F^- > Cl^- > Br^- > NO_3^- > SO_4^{2-}$ which clearly states that calcination does not affect this behaviour and ionic radius is an influencing factor even without calcination.

Keywords: Calcination, Effluent treatment, Hydrotalcite, Mg-Al LDH

Industrial effluents now stand as a major contributor to the destruction of river ecosystems. It contains many toxic as well as undesirable constituents such as heavy metals, oil, grease, organic compounds and even coarse and suspended particles. The chronic effects of this destruction not only disturb the ecological balance but also add pace to the drinking water pollution crisis.

One among the major problem we are facing today is the alarming increase of anionic species (such as fluoride, bromide, chloride, nitrate and sulphate) in water bodies. It is mainly caused due to effluents from various industries^{1,2}. Even if it is considered to be non-toxic to human beings, its increase can cause problems to aquatic ecosystems³. The increase in anionic nutrients will trigger and favour the abnormal growth of algae and weeds. This growth will result in severe depletion of dissolved oxygen in the water and reduce the oxygen availability to other aquatic organisms. Eventually, it results in the mass death of organisms which in turn causes pollution and reduction in the water quality of river bodies^{4,5}. Even though the legislation governing these emissions have strict rules and limits, these limits seem to be inadequate in facing this issue. Hence designing the safest method to remove various anions from effluent

water is an important task. Thus the development of an economically viable as well as time-bound method which easily blends in with the existing effluent purification methods is an important task. At the same time, this work is tedious since no existing specialised anionic species removal methods are used in industries today which makes this work novel and socially relevant.

Anionic species

Anions are ionic species with a negative charge. They may be a single atom or a group of atoms which are typically larger than cations (positively charged ions) due to the presence of extra electrons around them. The anions under consideration in this work are fluoride, bromide, chloride, nitrate and sulphate.

Fluoride at low concentrations is an essential element and it is even purposefully added to drinking water or table salt in low concentrations to protect against tooth cavities and decay. While chronic exposure to high concentrations of fluoride can lead to many health issues such as mottling of teeth and even skeletal fluorosis⁶. Nitrate is the main ingredient in fertilizers and also a part of the human diet as it is present in cured meat, leafy vegetables and even in drinking water⁷. Nitrate plays an important role in the

human body in reducing hypertension. While its excessive presence can lead to nitrate toxicity in infants which is often referred to as Blue baby syndrome. Excessive nitrate levels can result in Methemoglobinemia in pregnant women. The USEPA restricts the maximum allowable nitrate in drinking water to concentration to 10 ppm. Chlorides are the integral constituent of the human diet as it is present in Sodium chloride or commonly referred to as table salt. Making use of non-reactive solute property, chloride is used as an indicator of faecal contamination in river and groundwater. But the excessive levels of chlorine can lead to the severe body condition of hyperchloremia in human beings. This may cause a high level of dehydration and can affect the working of kidneys. Bromides are abundantly present in seafood and are part of the human diet but at the same time excess levels of bromide can cause lots of adverse effects to human health which includes restlessness, irritability, nausea, constipation and also many skin diseases like rashes acneiform etc. Sulphates are also part of the human diet as glucosamine sulphate which has an important role building tendons, cartilage, ligaments and the thick fluid that surrounds joints⁸. Excessive sulphate levels can cause catharsis, dehydration and even it can cause a severe case of diarrhoea.

It is a fact that anionic species are required in small amounts for human beings in the diet, but their abnormal concentration is under no circumstances acceptable and recommended at all.

Anionic contaminants are non-carcinogenic to human beings, non-lethal and non-mutagenic. This is why the industry gives less priority to reduce the level of anionic waste in its effluents. Inadequate

regulations and updated permissible limits regulating the release of industrial waste do not provide the industry with appropriate guidelines for the installation of advanced mechanisms or methods for the removal of anionic waste. At the same time, many ecological imbalances may be triggered by a rise in the number of anionic pollutants in effluents which can also contribute to a decrease in water quality. This not only leads to a lack of drinking water but also leads to limits on the use and consumption of fish and other marine species. In turn, this would cause the river and its habitat to be poisoned on a wide scale and ecologically ruined. For these purposes, similar to other priority contaminants, the removal of anionic waste from industrial effluents should be given equal significance. This includes the need to adequately analyze and prepare recommendations for the removal of anionic pollutants from industrial waste.

Layered double hydroxide

Layered double hydroxide (LDH) (also called as hydrotalcite) is one among the prime focus of research. The discovery of hydrotalcite backs to 1842 in Sweden, which was found to be an alloy of magnesium and aluminium^{9,10}. LDH (Fig. 1) can be easily prepared and it has numerous applications in catalysis, polymer additives, medical, wastewater remediation etc^{9,11}.

They are known to have the general formula $[M_{1-x}^{2+}M_x^{3+}(OH)_2]^{x+}[A^{n-}]_{x/n}.mH_2O$, in which M^{3+} and M^{2+} are cation layers with valency 3 and 2 respectively, the value of x lies between 0.2 and 0.33 and A^{n-} denotes anion which is exchangeable¹²⁻¹⁴. Hydrotalcite is an attractive mineral because it can be synthesized effortlessly and has applications in various fields¹⁵.

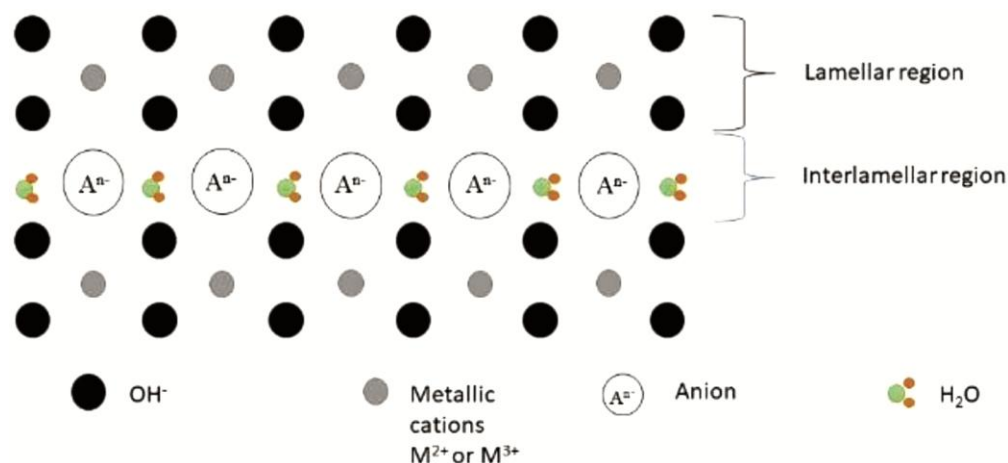


Fig. 1 — Structure of Hydrotalcite

This work evaluates the use of LDH in water treatment. The flexibility of hydrotalcite's structures and composition made them promising adsorbents to extract different hazardous pollutants from water bodies (Liang *et al.*, 2013; Yang *et al.*, 2016a,b)¹⁶⁻¹⁸. Such nanostructured hydrotalcite can therefore be used, primarily via adsorption and ion exchange mechanisms, to remove contaminants from wastewater (Gu *et al.*, 2015; El Hassani *et al.*, 2017; Pérez *et al.*, 2017; dos Santos *et al.*, 2017; Zubair *et al.*, 2017)¹⁹⁻²³. Pollutant adsorption on hydrotalcite is an efficient, economical, and non-toxic approach (Goh *et al.*, 2008; Liang *et al.*, 2013)^{16,24}. The combination of hydrotalcite with materials such as nanostructures of carbon, surfactants, nanoparticles of iron, and polymers showed major improvements in surface characteristics and adsorption performance (Mukkaram *et al.*, 2017)²⁵. The earlier studies addressed the adsorption efficiency of hydrotalcite-containing hybrids with respect to parameters such as pH, contact time, adsorbent dosage, and temperature for the removal of heavy metals and dyes (Mukkaram *et al.*, 2017)²⁵. Indeed, these hydrotalcite-containing hybrids have increasingly diverted interest in investigating their adsorption ability to remove hazardous pollutants from various aqueous solutions. The earlier reviews on hydrotalcite emphasized their methodologies of synthesis (Rives and Ulibarri, 1999; Goh *et al.*, 2008; Gu *et al.*, 2015)^{19,24,26} and application in catalysis and environmental contamination (Xu *et al.*, 2011; Fan *et al.*, 2014; Li *et al.*, 2014; Yang *et al.*, 2016)^{17,27-29}. Currently, a few techniques are available for the removal of anionic contaminants from aqueous solutions. It extends from simpler methods such as the use of red mud (Yunus *et al.*, 2002)³⁰, alum slum (Sujana *et al.*, 1998)³¹, quick lime (Islam *et al.*, 2007)³² etc to sophisticated electrochemical methods (Pulkka *et al.*, 2014)³³ and membrane techniques (Bodzek *et al.*, 2011)³⁴. Recent literature shows much eco-friendlier removal techniques. Use of Biochars is considered to be an eco-friendly removal method of anionic contaminants (Li *et al.*, 2019)³⁵. The use of Zn-Al LDH can be administered in the removal of methyl orange and Chromium anionic contaminants (Meng *et al.*, 2018)³⁶. While recent advancements also show light to the use of more biocompatible ternary Layered Double hydroxide for the removal of anionic organic dyes (Rathee *et al.*, 2019)³⁷. But no literature is found stating the direct use of Mg-Al

hydrotalcite in the removal of anionic contaminants for aqueous solutions.

Despite its inability in the selectivity of ions, hydrotalcite still serves as a sorbent in anion removal from aqueous solutions via the mechanisms of anion exchange and reformation effect. Even if anion exchange occurs in the natural state of hydrotalcite, better and superior results are found to be obtained on calcination (Miyata *et al.*, 1983; Lv *et al.*, 2006; Mokhtar *et al.*, 2012)³⁸⁻⁴⁰. The earlier studies reported that increasing the calcination temperature on hydrotalcite shows a positive trend towards the increase in its surface area (Julianti *et al.*, 2017)¹⁵. The reformation effect comes only on calcination. Reformation effect or memory effect is an unusual property shown by thermally activated LDH, in which it reforms back to its initial structure when exposed to any aqueous solutions (Olfs *et al.*, 2009; Mokhtar *et al.*, 2012)^{40,41}. This property is obtained via the loss of anions and interlayer water during calcination which causes mixed metal oxide formation (Lv *et al.*, 2006; Othman *et al.*, 2009; Palmer *et al.*, 2009)^{39,42,43}. Hence the present work emphasis on the impact of calcination on the anion removal capacity of hydrotalcite by reformation effect.

Mechanism

Thermally activated LDH shows an unusual property of reforming back to its initial structure when exposed to any aqueous solutions. This reformation occurs via the sorption of anions and water and is referred to as reformation or memory effect^{39,41}. To maximise this memory effect we can use calcination treatment. Calcination is performed by heat treatment of LDH, which results in the removal of anions and water trapped between the layers. This removal further results in a mixed metal oxide formation³⁸. Calcination mainly aims in the removal of the maximum number of anions possible creating more space to accommodate the desirable anions which we wish to remove from effluents. So care should be taken in choosing the anions initially during the hydrotalcite preparation in such a way that it can be easily removed at elevated temperatures by the formation of stable non-toxic decomposition products. In this work, carbonate is used as it can be easily prepared and during the removal, it decomposes into carbon dioxide and water vapour. The evolved gases are non-toxic at lower concentrations^{40,42,43}.

The basic mechanism of anion removal using Mg-Al hydrotalcite is sorption^{14,39-42}. Sorption is a combination of adsorption and absorption. Adsorption increases with the increase in surface area, while absorption increases in the increase in pore diameter and pore volume. Hence study about these parameters can help in predicting the mechanism of anionic species removal in calcinated and non-calcinated hydrotalcite. The optimum sorption efficiency of LDH mainly depends on two factors. 1) The maximum removal of interlayer anions as it will increase the efficiency and thus enhances the better performance of thermally activated LDH anions from LDH by calcination procedure and 2) selection of suitable calcination temperature^{14,40}. Exposure of LDH to excessive-high temperature can result in the partial decomposition of LDH structure leading to the formation of oxides which in turn obstructs the reformation behaviour of LDH. This causes a reduction of anion sorption efficiency which makes LDH unusable¹⁴. Hence the calcination temperature is a very important factor and must be considered carefully. The optimum calcination is determined experimentally since it varies for each LDH depending on its components. It is generally ranged between 300 and 800°C depending on the precursors used in LDH preparation^{40,41}. However, we chose the activation temperature range maximum of upto 500°C in this work. With the calcination treatment, we expect to increase the LDH surface area, pore diameter and pore volume which favours the increase in sorption of anions to LDH.

Experimental Section

Materials

Mg(NO₃)₂.6H₂O (Merck, 99%) and Al(NO₃)₃.9H₂O (Merck, 99%) were used for the precursor solution. For anionic solution, NaOH (Merck, 99%) and Na₂CO₃ (Merck, 99,9%) were used. For the preparation of anionic stock solutions Na₂SO₄ (Merck, 99%), NaCl (Merck, 99%), NaBr (Sigma Aldrich, 99%), NaNO₃.H₂O (Merck, 99%) and NaF (Merck, 99%) were used. All the reagents used were of analytical grade.

Methods

Synthesis of hydrotalcite

Several methods are available for the preparation of LDH such as the alkali metal method⁴⁴, the urea

method^{14,44}, the sol-gel method⁴⁴, the microwave irradiation method⁴⁴, The co-precipitation method⁴⁴ etc. Each method contributes to its own merits and demerits. Among the methods^{14,44,45}, the co-precipitation method is simple and most common. Hence we followed this method in this work. This method involves the preparation of a precursor solution and an anionic solution. The precursor solution contains desired cationic salts dissolved in Millipore water in a desired stoichiometric ratio. The anionic solution should have pH 9 or greater which contains the desired interlayer anion depending upon the methods chosen. The precursors used in this work are Mg(NO₃)₂.6H₂O and Al(NO₃)₃.9H₂O in which the Al/Mg molar ratio was taken as 3:1. The anionic solution was prepared by dissolving Na₂CO₃ in Millipore water by keeping the molar ratio of CO₃/(Mg+Al) as 0.66³⁸. Constant pH of 9 was maintained by adding the caustic solution. LDH was then prepared by adding the anionic solution dropwise into the precursor solution using a burette. The addition was done with vigorous mixing at 65°C. After adding the anionic solution completely, the mixture was kept undisturbed for 5 hours under the same condition of mixing and temperature. The precipitate formed was washed using distilled water until the pH reached 7 and then was separated using centrifugation. The separated precipitate was then kept for drying at 100°C overnight to remove excess water. The dried precipitate was crushed using a mortar and pestle and was uniformly sieved.

Treatment procedure

Calcination treatment was done in batches using a Muffle furnace. The procedure for calcination was adopted from literature (Julianti *et al.*, 2017)¹⁵. About 2g of Hydrotalcite was taken and subjected to different temperatures of 100, 200, 300, 400 and 500°C for 5 h.

Preparation of anionic stock solutions

The anionic stock solutions of Sulphate, Nitrate and Fluoride of 1000 ppm each was prepared by dissolving Na₂SO₄, NaNO₃, NaCl, NaBr and NaF in Millipore water respectively. The stock solution is further diluted to 50 ppm for experimental studies. Each sample of LDH was treated with 20 mL of each of the stock solutions separately. The mixture was taken in a conical flask and kept for uniform stirring using a shaking incubator for 3 h at 25°C. After stirring the LDH is filtered and the filtrate is analysed.

Characterisation and analysis

Hydrotalcite characterisation study was performed by SEM (Scanning Electron microscopy) and BET (Brunauer-Emmett-Teller). SEM was used to study the surface structure. The surface area investigation was done by the BET method. The anionic species quantification was performed using ion chromatography.

X-ray Diffraction analysis

Rigaku Miniflex 600 X-Ray Diffractometer was used for conducting X-ray diffraction analysis using Cu-K α radiation (0.154 nm). The scanning angle was from 3° to 90° and the scanning rate selected was 0.02 deg/s.

Morphological analysis

The morphological, as well as structural analysis of the Mg-Al hydrotalcite, were performed using Field Emission Scanning Electron Microscope model JEOL Model JEM 2100.

Thermogravimetric analysis (TGA)

The thermal stability of Mg-Al hydrotalcite was determined using Hitachi STA 7200 Thermal Analysis System. Mass loss corresponding to the temperature and thermal degradation pattern was obtained from TGA curves.

Surface area analysis

Brunauer Emmett Teller (BET) was used to study gas molecule adsorption on the solid surface. The quantity of gas adsorbed is highly related to the solid surface area⁴⁵. The surface area was analysed using MicrotracBEL Corp BELSORP Max. The adsorption and desorption experiments were conducted using gaseous nitrogen and temperature was controlled by using liquid nitrogen. Various parameters such as pore volume and average pore diameter were also analysed.

Anionic species quantification

The quantification of anionic species was done using ion chromatography. The analysis was done using the Metrohm 883 Basic IC Plus. For elutriation sulphuric acid was used as cation and the mixture of sodium carbonate and sodium bicarbonate was used as the anion. Millipore water is used as a neutral agent for optimisation of the column.

Results and Discussion

X-ray Diffraction analysis of hydrotalcite

X-Ray Diffraction analysis of synthesised Mg-Al hydrotalcite was conducted. The diffractogram

shows sharp peaks at low 2 θ angles but gives broad peaks at high 2 θ angles. The results obtained was analysed using X'Pert High Score Plus and were compared with (JCPDS) 00-022-0700 which has diffractogram peaks at 2 theta angles 11.277°, 22.783°, 34.467°, 38.610°, 46.284°, 59.983°, 61.891°. The diffractogram of synthesised hydrotalcite shows resemblance with (JCPDS) 00-022-0700 by showing diffraction peaks appearing at the 2 theta values 11.59°, 22.87°, 34.75°, 39.21°, 46.13°, 60.53° and 61.77°.

The d-space values for each 2 theta values were calculated using the Bragg's equation, $d = \frac{n\lambda}{2 \sin \theta}$ where d is the coherence length of the crystallite size of MgO-like phase, λ is the wavelength of the light used and θ is the diffraction angle (15). The calculated d-space values for each 2 theta values are 7.6 Å, 3.8 Å, 2.5 Å, 2.3 Å, 1.9 Å, 1.5 Å and 1.4 Å respectively. The diffractogram also showed some peaks appearing in 2 theta values 22.19°, 29.35°, 31.13°, 35.35°, 38.59° shows the presence of sodium nitrate which is a by-product in the hydrotalcite synthesis. This presence of sodium nitrate is confirmed due to its resemblance with (JCPDS) 00-001-0840 observed when analysed using X'Pert High Score Plus. The diffractogram obtained with peaks around 60° corresponds to the synthesised hydrotalcite with an interlayer of carbonate anion and nitrate anion^{3,46}.

Morphology of hydrotalcite

Figure 2 shows the SEM image of the hydrotalcite. It gives the idea that the hydrotalcite is well-formed as an accumulation of nano-sized particle aggregates and is arranged in a way like stacked one above the other. The image confirms the crystalline behaviour of hydrotalcite and matches with the characteristic reflections obtained in diffraction analysis⁴⁷.

Thermogravimetric analysis

The TGA plot of hydrotalcite show a reduction of 30% weight of LDH on exposure to 500°C.

The stagewise thermal decomposition of hydrotalcite can be explained as; the loss of intermolecular water molecules due to calcination which occurs at approximately 200°C. This step is irreversible. From 200°C to 400°C the hydroxide ions and carbonate ions intercalate with the bound water which results in the formation of CO₂ leading to the

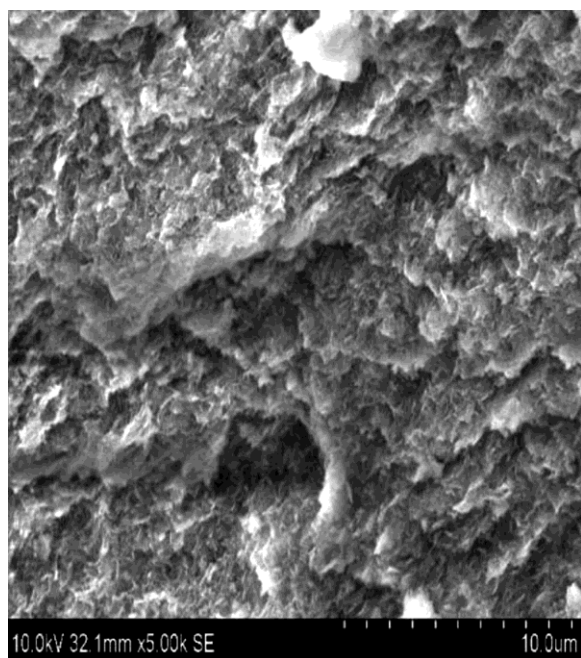


Fig. 2 — The SEM captured image of hydrotalcite

formation of $[Mg_{1-x}Al_xO(OH)_x]$. On further heating, from 400°C up to 500°C , the remaining hydroxide ions decompose which results in the formation of double oxides $[Mg_{1-x}Al_xO_{1+x/2}]^{15,43}$.

Calcination and mass loss

The calcination was performed batch-wise by exposing pre-weighed samples to different temperatures. The samples were calcinated for five hours. The percentage of mass loss shows a significant loss of water and other ions due to calcination. It shows that exposure to 500°C results in the loss of 30% of the initial mass of hydrotalcite taken.

BET analysis

The specific surface area found by BET method to the corresponding calcination temperatures shows an increasing behaviour. The cause of the increase in surface area can be explained as the crater (pore hole) production and decomposition of entire ion hydroxides¹⁵. The mean pore diameter and total pore volume show an increasing trend with the increase in calcination temperature. This trend is attributed due to the opening of pores in the calcinated hydrotalcite due to the removal of CO_3^{2-} from interlayer spaces²⁹.

Anionic species quantification

The filtrates of calcinated samples, as well as the non-calcinated sample, were analysed using Ion chromatography. The filtrate was analysed and the

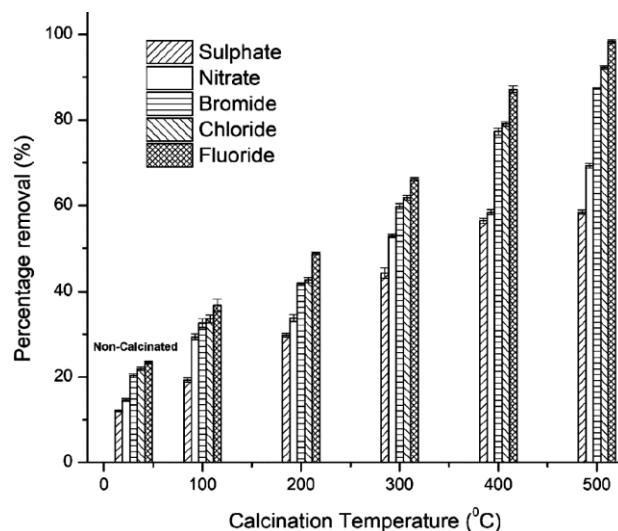


Fig. 3 — Percentage removal of various anions using non-calcinated and calcinated hydrotalcite

results were compared with the initially analysed blank solution. The removal efficiencies were calculated and are plotted in Fig. 3. Quantification was conducted three times and the obtained results are statistically confirmed by plotting Error bar Graph. The percentage of uncertainty is found to be minimal.

From the results shown in Fig. 3, the increase in calcination temperature shows an increasing trend towards the anionic removal capacity of hydrotalcite. It is observed that the maximum percentage of removal occurs in samples treated with hydrotalcite calcinated at 500°C and least for the samples treated with non-calcinated hydrotalcite. From the results, it is clear that calcination causes dehydration and this will increase disorders to the crystalline structure of hydrotalcite resulting in the formation of craters and potholes on its surface. The BET results also support this increase in the surface area along with the fact that the surface area, mean pore volume and mean pore diameter of the hydrotalcite increases with the increase in calcination temperature. This result suggests that calcination can cause an increase in surface area of hydrotalcite which favours adsorption. This provides a clear explanation for the increase of anionic removal capacity of hydrotalcite with the increase in calcination temperature.

Among the five anions studied, Mg-Al hydrotalcite shows maximum removal towards fluoride (98.4 %) and least towards sulphate (58.4 %). At the same time, the percentage removal of Chloride, Bromide and Nitrate is found to be 92.3%, 87.4% and 69.3%

respectively for the sample calcinated at 500°C. When non-calcinated hydrotalcite samples are used the percentage removal of anions are found to lower than the calcinated sample. However, in the case of the non-calcinated sample, the maximum percentage of removal was for fluoride (23.4 %) and minimum for sulphate (12.1 %). The percentage removal of Chloride, Bromide and Nitrate ions is found to be 21.9%, 20.3% and 14.6% respectively.

For both, calcinated and non- calcinated samples, it is found that the percentage removal of anions decreases with the increase of its ionic radii. i.e. $F^- > Cl^- > Br^- > NO_3^- > SO_4^{2-}$. This behaviour can be explained in such a way that, as the ionic radii increases, its size also increases and hence the number of anions which can be accommodated decreases. And calcination seems no effect on this behaviour which gives us a clear idea that the ionic radius is an influencing factor even without calcination.

Comparison with similar works

The higher removal values obtained in our work provides a confident and comparable result with existing works of literature. An earlier reported work shows 98% removal of fluoride from aqueous solution using Mg-Al hydrotalcite calcinated at 500°C (Lv *et al.*, 2006)³⁹. But our current results show a slight increase in the percentage removal of fluoride under similar condition. In the case of chloride current study shows a very high percentage removal value of 92.3% comparing to the earlier reported value of 64.2% using Zn-Al LDH as anion exchanger (Lv *et al.*, 2009)⁴⁸. The current percentage removal value of nitrate seems much higher when compared to an earlier result obtained in literature conducted to study the removal of nitrate ions using Mg-Fe modified wheat straw biochar. The earlier result obtained was 19.1% removal of nitrate ions by using 2 gm of biochar adsorbent (Xue *et al.*, 2016)⁴⁹. In this similar condition, Mg-Al hydrotalcite gave 69.35% of nitrate removal. The percentage removal of bromide obtained in this present work seems comparable and higher when compared to the result obtained in the literature studying the removal of bromide ion from aqueous solution using silver-impregnated activated carbon (Chen *et al.*, 2017)⁵⁰. The removal study using silver-impregnated activated carbon gave a percentage removal of 72% which is lesser when compared to 87.4% removal of bromide ions in the present work. However, in the case of sulphur, the percentage removal from the current study seems to be lesser

when compared to earlier reported literature which studied the sulphate ion removal using modified rice straw (Cao *et al.*, 2011)⁵¹. The earlier study gave 78.9% removal which is comparably higher to 58.4% removal in the current work. This reduction can be due to limitation of Mg-Al hydrotalcite in accommodating more number of sulphate ions which has large ionic radius and ionic size.

Comparing to anion removal techniques such as the use of biochars, hydrotalcite seems much easily preparable. And it also seems very cost-effective when comparing to electrochemical and membrane separation methods. However, the higher anionic removal values in the current work stand as a promising result to pursue more research in the same.

Conclusion

The study investigated the effect of calcination on various parameters such as surface area, mean pore diameter and total pore volume for the anion removal efficiency of Mg-Al hydrotalcite under reformation effect. These parameters show an increasing trend with the increase in calcination temperature. The formation of craters and potholes due to calcination increases the surface area of hydrotalcite which triggers the increase in adsorption and thereby increases the capacity for the removal of anionic species. The percentage removal of all anions under study increases with the increase in calcination temperature. At the same time, for both calcinated and non- calcinated samples, it is found that the percentage removal of anions decreases with the increase of its ionic radii as. $F^- > Cl^- > Br^- > NO_3^- > SO_4^{2-}$.

From the studies, it is concluded that calcination can be used as a method to increase the anionic removal efficiency of hydrotalcite from aqueous solutions. Calcination increases the surface area of hydrotalcite and thereby enhances the anionic species removal capacity of Mg-Al hydrotalcite. It is also observed that the percentage removal of anions decreases with the increase of its ionic radii and the ionic radius is an influencing factor even without calcination.

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