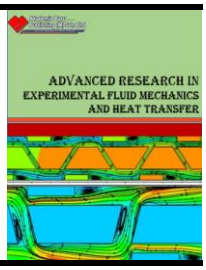
Penerbit Akademia Baru

Journal of Advanced Research in Experimental Fluid Mechanics and Heat Transfer

http://www.akademiabaru.com/submit/index.php/arefmht

ISSN: 2756-8202



Characterization and Thermal Behaviour of Magnesium-Aluminium Layered Double Hydroxide

Nurul Iman Abdul Razak[1], Noor Izyan Syazana Mohd Yusoff[2], Mat Uzir Wahit[1,3*]

[1] School of Chemical and Energy, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia
[2] Advanced Membrane Technology Research Centre (AMTEC), Universiti Teknologi Malaysia, 81310, Skudai, Johor, Malaysia
[3] Center for Advanced Composite Materials, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

ABSTRACT

Layered double hydroxide achieved a lot of attention because it is simple, eco-friendly and easy to synthesize, involved non-toxic chemical, economical, good thermal stability with broad spectrum application. Thus, layered double hydroxide gained trust by the academician and industrial in developing layered double hydroxide studies. Thermal behaviour does influence by layered double hydroxide molar ratio therefore this study focuses on the molar ratio effect of Mg-Al nitrate LDH on the characterization and thermal behaviour properties. The divalent and trivalent cation of magnesium nitrate hexahydrate (Mg $(NO_3)_2{\cdot}6H2O$) and aluminium nitrate nonahydrate (Al $(NO_3)_3{\cdot}9H2O$) ratio are 2:1, 3:1 and 4:1 synthesize by co-precipitation method. The X-ray diffraction analysis shows lower molar ratio obtain high crystallinity index as aluminium concentration decreased and the crystallinity does affect the heat flow of the material in differential scanning calorimetry analysis. A typical presence of the inorganic-organic compound within the network structure of layered double hydroxide in Fourier transform infrared spectroscopy. The thermal stability from thermogravimetric analysis proved decomposition occurred with formation of oxide mixture in the materials network.

Keywords:
Layered double hydroxide; co-precipitation; anionic clay;thermal behaviour; nano material

1. Introduction

In recent years, layered double hydroxide (LDH) has been received considerable attention due to their wide spectrum applications especially in polymer nanocomposite. LDH been known by many names such as hydrotalcite compound due to high water content and talc-like or known as anionic clay because of containing anions at interlayer galleries for charge balance. The LDH general formula is $[M^{2+}_{1-x}M_x^{3+}(OH)_2]^{x+}(A^{n-1})$ $x/n.yH_2O$ where M^{2+} and M^{3+} are divalent and trivalent metal cations in the sheet, respectively, x is the ratio of M^{3+}: $(M^{2+} + M^{3+})$ which divalent constructed from but not limited to Ca^{2+}, Co^{2+}, Cu^{2+}, Mg^{2+}, Ni^{2+}, Zn^{2+}, Mn^{2+} while trivalent Al^{3+}, Fe^{3+}, Co^{3+}, Mn^{3+}, Cr^{3+}. The A^{n-} is the interlayer anion where keep overall charge neutral such as CO_3^{2-}, NO_3^-, Cl^-, or SO_4^{2-} place at interlayer galleries. The one of natural LDH available is Mg_6 $Al_2(OH)_{16}CO_3.4H_2O$ but low purity thus LDH is preferable synthesize in laboratories with relatively low cost and high purity. The complex structure owned by LDH makes it has high thermal stability [1]. Mg and Al is most abundant and common element in earth, both materials have high thermal conductivity, low mass density and engineered coefficient of thermal expansion [2] thus Mg/Al is the most suitable materials which

required high thermal application. Besides, the molar ratio of the materials is the main key to obtain the best for thermal material. Interesting to note, divalent trivalent cation ratio must within 2:1 to 4:1 and will not form if it below or beyond the ratio, thus the study focuses within the ratios [3].

LDH can be prepared by numerous methods including co-precipitation, urea hydrolysis, the sol-gel method, hydrothermal synthesis, reformation and mechanical milling among others. Most common method used to prepare LDH is co-precipitation due to simple direct method, low-cost method with high yield production of LDH [4-6] thus it was the main focus in this study. The yield from co-precipitation method is three times more as compared to other routes and the particle size of LDH can obtain in nano sized between 60 to 80 nm [6]. Several parameters of this method such as pH of medium reaction, temperature, condition of metallic salts and anion species, concentration of alkali solution can be controlled independently. Conventional titration of alkali solution such as sodium hydroxide (NaOH), sodium carbonate (Na_2CO_3), potassium hydroxide (KOH) and ammonium hydroxide (NH_4OH) is added to aqueous solution containing M^{2+} and M^{3+} to desired pH and remain constant for the precipitation of targeted LDH.

LDH offer large surface area, layered crystalline geometry and ability to interchange interlayer anions with much larger organic anionic molecules. The layered structures of LDH helps induce interlayer spacing thus offer intercalation of polymer chain and offer interaction between filler and polymer. Due to the outstanding characteristics possessed by LDH, it has achieved lot of attention from academia and industries. The differences of LDH to other's clay namely montmorillonite such due to their composition, layer thickness and geometries on structural and chemical however the interlamellar region are similar to montmorillonite. LDH contains of anions in interlamellar as seen the hydroxide layers are stacked and held together bond by Vander Waal's force to stabilize the LDH structure while the divalent and trivalent metal hydroxide was bind with water molecule and anions with hydrogen bonding with positive charges, a single octahedral metal hydroxide crystal layer while montmorillonite is in opposite charges in two or more sheet of metal oxides [4][8] as is shown in Fig.1. Moreover, LDH has ability to reconstruction back to original structure when heated within 300–500 °C and LDH is not work in at acidic pH [3].

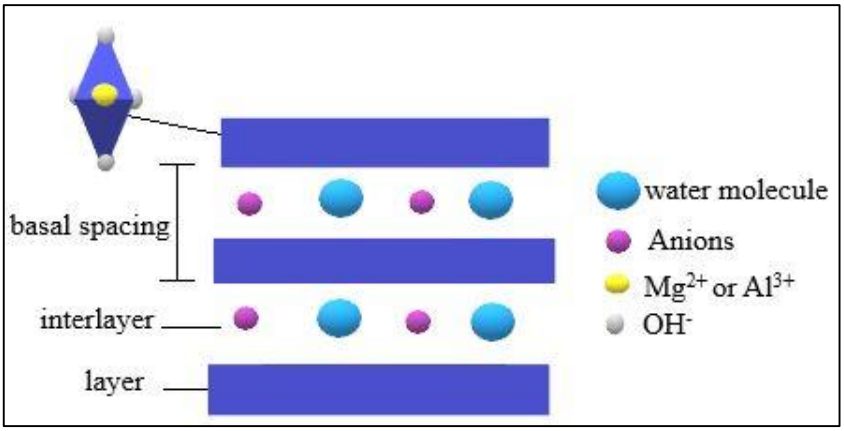


Fig. 1. Schematic diagram of Mg-Al nitrate layered double hydroxide

The typical white powder LDH able to reflect the heat and assist to improved heat insulation with aggregating on the surface and release water at 220 ◦C to 330 ◦C [9] thus clearly improved thermal stability which heat release rate is decreasing. Furthermore, LDH will improve aging resistance under both thermo-oxidative aging and ultraviolet (UV) aging by blocking the oxygen and reflecting the high-energy UV light at the interface of the LDH sheets respectively [10]. Moreover, LDH comes with

economical, involved non-toxicity materials, minimal corrosion, and excellent flammability behaviour. The properties possessed by LDH, it is widely used in many fields, included catalyst/adsorption, absorbents, electrical and electronic industries and even medical applications. In polymer additive industries, LDH act as fire-resistant coating which give advantages in a rust-proof and fire-resistant coating with a high film adhesion. The act as flame retardant also applied with phosphoric acid, fatty acid, aluminium hydroxide, magnesium hydroxide, magnesium hydroxide carbonate due to endothermic decomposition [11-12].

Various researchers have studied on thermal behaviour Mg/Al with molar ratio 2:1 and 3:1 which varies anionics such as chloride [13], carbonate [14], nitrate at 2:1 molar ratio using the urea hydrolysis method [15]. However up until now, the effect of Mg/Al nitrate LDH on thermal behaviour influences by the molar ratio range 2:1 to 4:1 has yet to be reported. Therefore, this paper presents the study of the thermal behaviour and characteristics of Mg/Al nitrate LDHs in three different molar ratios by co-precipitation method.

2. Methodology

2.1 Materials

Magnesium nitrate hexahydrate (Mg $(NO_3)_2 \cdot 6H_2O$) (99% A.C.S. reagent) and sodium hydroxide (NaOH) (97% reagent) will be obtained from Honeywell Fluka™. Aluminium nitrate nonahydrate (Al $(NO_3)_3 \cdot 9H_2O$) from Sigma Aldrich (M) Sdn. Bhd and sodium carbonate (Na_2CO_3) from Qrec (Asia) Sdn Bhd. All chemical is analytical reagent grade and used as received, without further purifications. Deionized water was used as solvent throughout the study.

2.2 Synthesis of LDH

Mg/Al nitrate LDH was synthesized by most reliable techniques co-precipitation method. The solution of $MgCl_2 \cdot 6H_2O$ and $AlCl_3 \cdot 6H_2O$ with different molar ratio were prepared (2:1, 3;1 and 4:1). The mixture solution of $MgCl_2 \cdot 6H_2O$ and $AlCl_3 \cdot 6H_2O$ was vigorously stirred for next 6 hours using magnetic stirrer at room temperature. Meantime, prepared NaOH solution 2 M and Na_2CO_3 solution 1 M under continuous stirring conditions and then the base mixture solution was added dropwise to the mixture of $MgCl_2 \cdot 6H_2O$ and $AlCl_3 \cdot 6H_2O$ solution. After the drop-wise addition was complete the slurry was aged at 65°C for 18 hours prior. During the reaction, pH needs to adjust and kept constant at pH 10. Then centrifuged to cover white solid at 5000 rpm for 5 minutes to separate the wet cake and washed with deionized water to remove excessive salts. The white precipitation was dry overnight at 100°C to obtain pure Mg/Al LDH powders.

2.4 Characterization of LDH

The LDH crystal formation was analysed with Rigaku SmartLab XRD, the pattern powdered sample 3 s per interval from 10°C to 80°C. From the data, the crystallinity index and d-spacing using Bragg's equation can be calculated. The FTIR spectra were recorded in a Perkin–Elmer 2000 spectrophotometer in the form of powder. The changing transmittance peak and/or intensity of functional groups in the IR spectra are using to identify the interaction of Mg/Al nitrate LDH. The characterization was carried out in the range of 4000–400 cm^{-1}. The result is supported with morphology analysis of the sample. The sample was examined with Hitachi HT7700 high resolution-transmission electron microscope (HR-TEM) at 1200 kV and were sputter coated with platinum before observation under TEM.

2.3 Thermal Testing

The thermal behaviour was obtained via Perkin Elmer thermogravimetric analysis (TGA) with a heating rate of 10°C/min from 25 to 900 °C under a nitrogen purge stream with the nitrogen flow rate of 50 mL/min and differential scanning calorimetric (DSC) analyses perform via Mettler Toledo DSC1 STAR system. The samples were weighed about 5-10 mg and sealed in an aluminium pan. The heating and cooling rate were 10 °C min^{-1} within temperature range of 25 to 550 °C. The characterization was conducted under nitrogen atmosphere with flow rate at 5.0 ml min^{-1}.

3. Results

3.1 XRD Analysis

The crystallinity of Mg/Al LDH is identified by XRD analysis and it is shows typical reflection peaks are present with Mg^{2+} and Al^{3+} cations structure in the inorganic sheets and balanced with NO^{-}_{3} at the interlayer spacing. The intensity is related with Mg/Al LDH crystallinity which the intensity decreases and the peak gradually broadens as the Al content decreasing. The reasons are large different of ionic radii between Mg^{2+} and Al^{3+} metal cation cause the distortion of the OH- layers networks [15] and the large polarization of Al^{3+} in layered structure thus the difficulties in directional alignment occurred [16]. Fig. 2 shows 8 diffraction peaks were detected at 2θ angle of 11°, 23°, 29°, 35°, 39°, 47°, 60° and 62° present in each molar ratio and the peak pattern consists of both asymmetrical and symmetrical. The sharpened peak has good crystallinity was obtained at molar ratio 2:1 with crystallinity index 87% followed by 78% and 67% for 3:1 and 4:1 respectively. The bonding stability is increase as decreasing the molar ratio and it has good agreement with literature [17] the crystallinity of materials increase when molar ratio decreases. The results obtain is in good agreement with these literatures, a significant increase of crystallinity in decreasing molar ratio 4:1 <3:1 < 2:1.

The d-spacing calculated are 7.72 Å, 7.78 Å and 7. 80 Å as increase the molar ratio 2:1, 3:1 and 4:1 respectively. There is discussion among scholar which basal spacing or d-value is perpendicular with the vertical or horizontal orientation interlayer or it is due to changing of layer charge density resulted from configuration of a single flat-lying molecular layer to multiple layers in LDH interlayers. The 2:1 molar ratio the d-value is 7.72 Å which the smallest basal spacing and it is suggested the interlayer in horizontal orientation or no significant changing of charge density while 3:1 and 4:1 has bigger basal spacing and both has similar value 7.78 Å and 7. 80 Å respectively. It is suggested both are in vertical orientation and charge density is happening [18].

The composition of LDH is confirm by TEM EDX and the image is platelet-like particle is shows in Figure 3 spectrum. Refer to schematic diagram in Fig. 1, most element present in LDH is oxygen with 59 wt.% including part of water molecule, anionic interlayer and some are bonding with metal oxide. As can see the ratio Mg to Al is 14.2 to 4.9 and interlayer anions element is 4.6 wt.%.

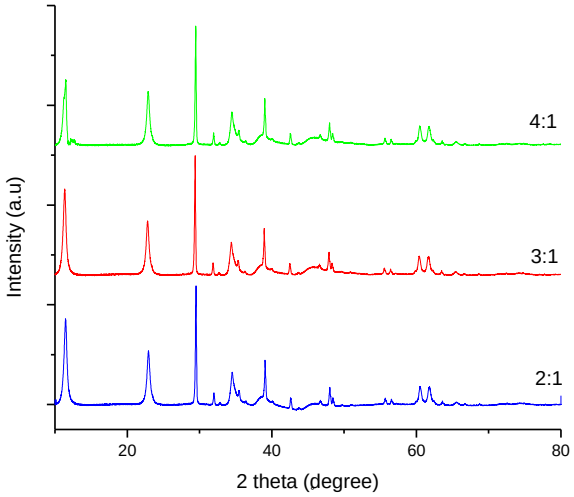


Fig.2. XRD analysis of 2:1, 3:1 and 4:1 Mg-Al nitrate layered double hydroxide

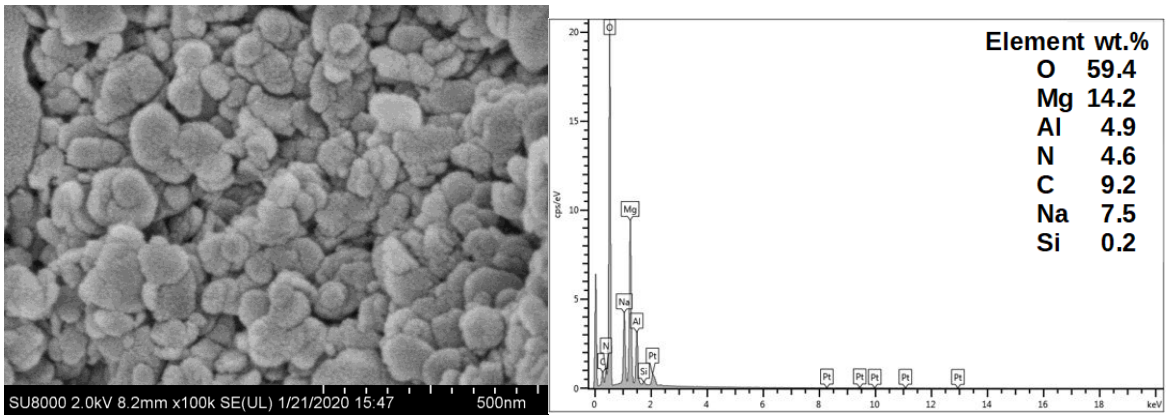


Fig. 3. TEM morphology and composition elements of Mg-Al nitrate layered double hydroxide

3.2 FTIR

Figure 4 illustrates the FTIR spectra at different LDH molar ratio shows similar types of features. Broad absorption band at 3466 cm^{-1} is attributed to the stretching vibration of -OH in the brucite-like layers and the interlamellar water molecules. The free state of -OH is being shifted towards low wave number and strongly suggested the presence of -OH bonds between -OH layer and interlayer water. The low intensity band at 1638 cm^{-1} are attributed to the bending vibration mode of the Mg/Al nitrate LDH interlayer–water interaction or called solvation. The sharp absorption peak at 1384 cm^{-1} is the bending vibration of metal hydroxide sheets and the stretching vibration of NO_3^- in the Mg/Al nitrate LDH interlayer. The bands observed around 839 and 670 cm^{-1} attributed to out-of-plane symmetric deformation mode and the anti-symmetric deformation mode of nitrate. The absorption peaks around 770–550 cm^{-1} correspond to the lattice vibration of metal–oxygen bonds (Mg–O and Al-O) stretching modes. [19-22].

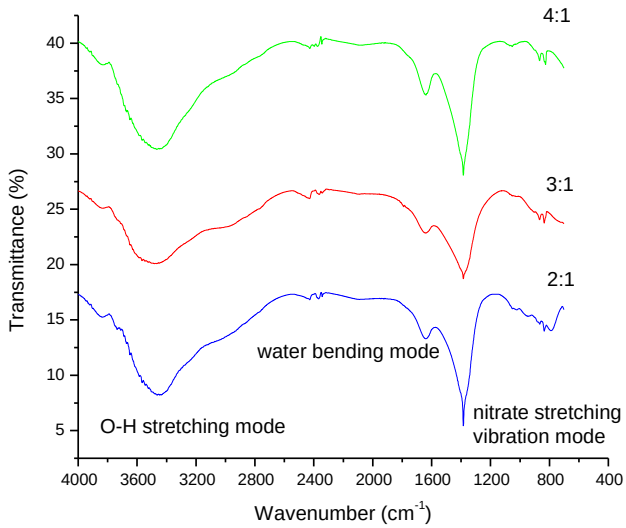


Fig.4. FTIR spectrum of Mg-Al nitrate layered double hydroxide at different molar ratio

3.3 Thermal Behaviour

The thermal behaviour of Mg/Al nitrate LDH was examined via TGA and DSC. According to results obtain in Fig 5, shows the curves trend is typical for hydrotalcite structure. The relation between thermal conductivity and molar ratio in this study is leaving much desired as much known decreased Mg/Al LDH crystallinity as Mg/Al molar ratio increase will be decreasing thermal conductivity, however this can be explained which may result from the random disorder phonon scattering induced by the Al deficient sites [16]. The TGA and DTG curves shows three decomposition stages accompanied by endothermic of Mg/Al LDH and the first stage called low-temperature decomposition. The strong interactions between interlayer water molecules and interlayer NO_3^- helps in delay the loss of water molecules. The researchers agreed that temperature around 200°C is attributed to the loss of water and interlayer crystallization water molecules [23-24]. Here, the amount of the free water and interlayer water molecules about 7 wt.%.

The second decomposition step known as dehydroxylation of OH- bonded with Mg^{2+} or Al^{3+} and the beginning of interlayer anions degradation [13]. The mass losses are 15wt.% occurred at temperature <330°C to decompose. The decomposition of interlayer anions is just about to begin at these temperature ranges and the LDH structure collapsed at 350°C [25]. Wang et.al suggests that the NO_3^- anions decompose only after the destruction of the lamella structure of LDH [26]. In third step decomposition obviously see the NO_3^- anions decomposition occurred with the mass loss region 34wt.% at temperature range about 600°C. The char residue values for all samples are about 45 wt.%.

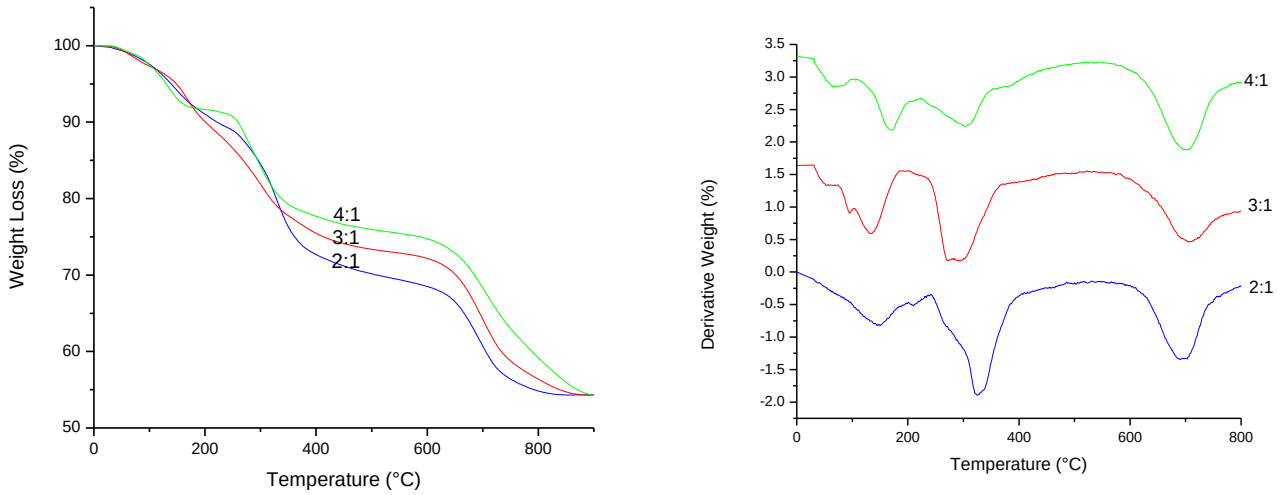


Fig. 5. TGA and DTG curves of Mg-Al nitrate layered double hydroxide at different molar ratio

Fig. 6 shows DSC analysis of Mg/Al LDH and the trend of DTG curve have good agreement with DSC measurement which has three steps decomposition and enthalpy. The first curve entitles to water losses were below 200°C. The next curve shows tighter and stronger bond thus harder to release the OH^- from Mg^{2+} and Al^{3+} where is the molar ratio effect the heat flow as Mg increase more heat is required due to magnesium has high polarization then aluminium. Thus, further heating is needed to decomposed the molecule [23,27] and it shows at molar ratio 2:1 with enthalpy 448 J/g followed by 419 and 247 J/g to 3:1 and 4:1 respectively. The formation of oxides and the anions decomposition [25] need more heat to breakdown the bond at molar ratio 2:1. It has agreement with XRD analysis which lower molar ratio has good crystallinity structure form of Mg/Al LDH due to increasing of Mg content. For the last DSC measurement, it shows different molar ratio has no significant effect on decomposition of anionic decomposition due to all of them as same anionic interlayer, NO_3^- the heat flow is 540, 526 and 516 J/g.

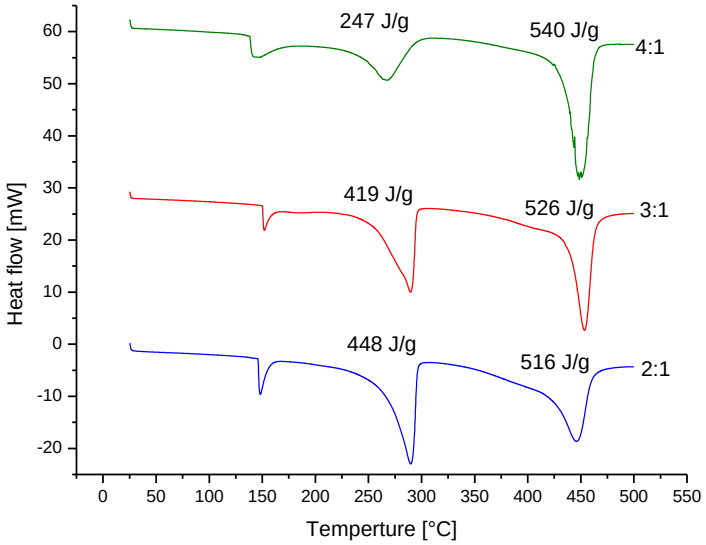


Fig.6. DSC curves of Mg-Al nitrate layered double hydroxide at different molar ratio

4. Conclusions

The Mg/Al LDH synthesized by co-precipitation method with molar ratio 2:1 has better bonding stability thus it is the best crystallinity structure by XRD and composition confirm by TEM EDX. The crystallinity structure with further heating is required to breakdown the bond which has strong interaction between two elements via DSC measurement. Thermal stability at higher molar ratio is obtained due to random disorder phonon scattering induced by the Al deficient sites. While the decomposition of anionic has no effect at different molar ratio due to all samples are NO_3^- anions.

Acknowledgement

This study was supported by UTM Fundamental Research Grant no: Q.J130000.2551.20H68, Reference no: PY/2019/01755

References

[1] dos Santos, Grazielle Emanuella de Souza, Pollyanna Vanessa dos Santos Lins, Leonardo Mendonça Tenório de Magalhães Oliveira, Elenara Oliveira da Silva, Ioannis Anastopoulos, Alessandro Erto, Dimitrios A. Giannakoudakis, André Ricardo Felkl de Almeida, Jose Leandro da Silva Duarte, and Lucas Meili. "Layered double hydroxides/biochar composites as adsorbents for water remediation applications: Recent trends and perspectives." *Journal of Cleaner Production* 284 (2021): 124755.

[2] Oddone, Valerio, Benji Boerner, and Stephanie Reich. "Composites of aluminum alloy and magnesium alloy with graphite showing low thermal expansion and high specific thermal conductivity." *Science and Technology of advanced MaTerialS* 18, no. 1 (2017): 180-186.

[3] Mishra, Geetanjali, Barsha Dash, and Sony Pandey. "Layered double hydroxides: A brief review from fundamentals to application as evolving biomaterials." *Applied Clay Science* 153 (2018): 172-186.

[4] Daniel, Saju, and Sabu Thomas. "Layered double hydroxides: fundamentals to applications." In *Layered Double Hydroxide Polymer Nanocomposites*, pp. 1-76. Woodhead Publishing, 2020.

[5] Mallakpour, Shadpour, and Elham Khadem. "Opportunities and challenges in the use of layered double hydroxide to produce hybrid polymer composites." In *Hybrid Polymer Composite Materials*, pp. 235-261. Woodhead Publishing, 2017.

[6] Theiss, Frederick L., Godwin A. Ayoko, and Ray L. Frost. "Synthesis of layered double hydroxides containing Mg2+, Zn2+, Ca2+ and Al3+ layer cations by co-precipitation methods—A review." *Applied Surface Science* 383 (2016): 200-213.

[7] Tonelli, Domenica, Isacco Gualandi, Elisa Musella, and Erika Scavetta. "Synthesis and Characterization of Layered Double Hydroxides as Materials for Electrocatalytic Applications." *Nanomaterials* 11, no. 3 (2021): 725.

[8] Wijitwongwan, Rattanawadee Ploy, Soontaree Grace Intasa-ard, and Makoto Ogawa. "Preparation of layered double hydroxides toward precisely designed hierarchical organization." *ChemEngineering* 3, no. 3 (2019): 68

[9] Rezvani Ghomi, Erfan, Fatemeh Khosravi, Zahra Mossayebi, Ali Saedi Ardahaei, Fatemeh Morshedi Dehaghi, Masoud Khorasani, Rasoul Esmaeely Neisiany et al. "The flame retardancy of polyethylene composites: From fundamental concepts to nanocomposites." *Molecules* 25, no. 21 (2020): 5157.

[10] Zhu, Kai, Yunhe Wang, Daquan Tang, Qiang Wang, Haihang Li, Yadong Huang, Zhiyi Huang, and Ke Wu. "Flame-retardant mechanism of layered double hydroxides in asphalt binder." *Materials* 12, no. 5 (2019): 801.

[11] Leroux, Fabrice, Thomas Stimpfling, and Horst Hintze-Bruening. "Relevance and performance of LDH platelets in coatings." *Recent patents on nanotechnology* 6, no. 3 (2012): 238-248.

[12] Mochane, Mokgaotsa Jonas, Sifiso Innocent Magagula, Jeremia Shale Sefadi, Emmanuel Rotimi Sadiku, and Teboho Clement Mokhena. "Morphology, thermal stability, and flammability properties of polymer-layered double hydroxide (LDH) nanocomposites: A Review." *Crystals* 10, no. 7 (2020): 612.

[32] Zhang, Yuze, Lijuan Li, Dong Shi, and Fugen Song. "Synthesis and application of low-cost layered double hydroxides intercalated by gluconic acid anion for flame retardancy and tensile strength conservation of high filling epoxy resin." *Journal of Colloid and Interface Science* 594 (2021): 791-801.

[14] Zhitova, Elena S., H. Chris Greenwell, Mariya G. Krzhizhanovskaya, David C. Apperley, Igor V. Pekov, and Victor N. Yakovenchuk. "Thermal Evolution of Natural Layered Double Hydroxides: Insight from Quintinite, Hydrotalcite, Stichtite, and Iowaite as Reference Samples for CO3-and Cl-Members of the Hydrotalcite Supergroup." *Minerals* 10, no. 11 (2020): 961.

[15] Quispe-Dominguez, Roger, Sajid Naseem, Andreas Leuteritz, and Ines Kuehnert. "Synthesis and characterization of MgAl-DBS LDH/PLA composite by sonication-assisted masterbatch (SAM) melt mixing method." *RSC advances* 9, no. 2 (2019): 658-667.

[16] Heraldy, E., K. D. Nugrahaningtyas, F. B. Sanjaya, A. A. Darojat, D. S. Handayani, and Y. Hidayat. "Effect of reaction time and (Ca+ Mg)/Al molar ratios on crystallinity of Ca-Mg-Al layered double Hydroxide." In *IOP Conference Series: Materials Science and Engineering*, vol. 107, no. 1, p. 012025. IOP Publishing, 2016.

[17] Ahmed, Abdullah Ahmed Ali, Zainal Abidin Talib, and Mohd Zobir Hussein. "Influence of metallic molar ratio on the electron spin resonance and thermal diffusivity of Zn–Al layered double hydroxide." *Journal of Nanomaterials* 2013 (2013).

[18] Elhalil, Alaâeddine, Meryem Farnane, Aicha Machrouhi, Fatima Zahra Mahjoubi, Rachid Elmoubarki, Hanane Tounsadi, Mohamed Abdennouri, and Noureddine Barka. "Effects of molar ratio and calcination temperature on the adsorption performance of Zn/Al layered double hydroxide nanoparticles in the removal of pharmaceutical pollutants." *Journal of Science: Advanced Materials and Devices* 3, no. 2 (2018): 188-195.

[19] Ayawei, Nimibofa, Seimokumo Samuel Angaye, Donbebe Wankasi, and Ezekiel Dixon Dikio. "Synthesis, characterization and application of Mg/Al layered double hydroxide for the degradation of congo red in aqueous solution." *Open Journal of Physical Chemistry* 5, no. 03 (2015): 56.

[20] Dasgupta, Sudip. "Controlled release of ibuprofen using Mg Al LDH nano carrier." In *IOP Conference Series: Materials Science and Engineering*, vol. 225, no. 1, p. 012005. IOP Publishing, 2017.

[21] Iqbal, Muhammad Ahsan, and Michele Fedel. "Effect of synthesis conditions on the controlled growth of MgAl–LDH corrosion resistance film: structure and corrosion resistance properties." *Coatings* 9, no. 1 (2019): 30.

[22] Seidi, Farzad, Maryam Jouyandeh, Seyed Mohammad Reza Paran, Amin Esmaeili, Zohre Karami, Sébastien Livi, Sajjad Habibzadeh, Henri Vahabi, Mohammad Reza Ganjali, and Mohammad Reza Saeb. "Imidazole-functionalized nitrogen-rich Mg-Al-CO3 layered double hydroxide for developing highly crosslinkable epoxy with high thermal and mechanical properties." *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 611 (2021): 125826.

[23] Golovin, Sergei N., Maksim N. Yapryntsev, Irina G. Ryl'tsova, Sergey V. Savilov, Konstantin I. Maslakov, and Olga E. Lebedeva. "Synthesis and thermal behavior of Co/AlCe layered double hydroxide." *Solid State Sciences* 111 (2021): 106498.

[24] Li, Tianxiang, Zhengren Shi, Xianru He, Ping Jiang, Xiaobin Lu, Rui Zhang, and Xin Wang. "Aging-resistant functionalized ldh–sas/nitrile-butadiene rubber composites: preparation and study of aging kinetics/anti-aging mechanism." *Materials* 11, no. 5 (2018): 836.

[25] Bujdosó, Tamás, Ágnes Patzkó, Zoltán Galbács, and Imre Dékány. "Structural characterization of arsenate ion exchanged MgAl-layered double hydroxide." *Applied clay science* 44, no. 1-2 (2009): 75-82.

[26] Wang, Liren, Zhi Lü, Feng Li, and Xue Duan. "Study on the mechanism and kinetics of the thermal decomposition of Ni/Al layered double hydroxide nitrate." *Industrial & engineering chemistry research* 47, no. 19 (2008): 7211-7218.

[27] Mahjoubi, Fatima Zahra, Abderrahim Khalidi, Mohamed Abdennouri, and Noureddine Barka. "Zn–Al layered double hydroxides intercalated with carbonate, nitrate, chloride and sulphate ions: Synthesis, characterisation and dye removal properties." *Journal of Taibah University for Science* 11, no. 1 (2017): 90-100.