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Preparation and analysis of multi-layered hybrid nanostructures

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Abstract

In this study, we report a simple and efficient (bath ultrasonication) method for the preparation of multi-functional layered hybrid nanostructures using graphene oxide (GO) and modified Mg-Al layered double hydroxides (LDH) [LDH@GO]. The functional composition, crystalline nature, layered surface morphology and thermal behavior of the hybrid nanostructures of LDH@GO were characterized by Fourier transform infrared spectroscopy, X-ray diffraction, Field-emission scanning electron microscopy, Transmission electron microscopy, elemental analysis and thermo gravimetric analysis. The results confirmed the formation of layered hybrid nanostructures of LDH@GO showing a significant increment in the spacing between GO sheets due to the incorporation of LDH. Also hybrid nanostructures of LDH@GO have better thermal stability as compared to pristine GO.

Keywords: Graphene oxide, Layered double hydroxide, Surface modification, Hybrid nanostructures, Properties.

1. Introduction

The field of material science and nanotechnology has blossomed over the last two decades due to various applications from medical to industrial sectors and from laboratory to market (Das and Prusty et al., 2013). But some of nanomaterials (NM)were observed with their aggregation due to the high specific area(Bourlinos et al., 2009; Mishra et al., 2010; Mali et al., 2012; Mishra et al., 2012; Esmaeili, et al.,2014; Angelopoulou et al., 2015) and also these NM were reported with specific use. On other side, alloying or hybridizing the two or more NM can give multifunctional properties of the resulting hybrid nanostructures (NS) and also they can be used in many applications. The combination of multi-dimensional NM or layered NS were used to prepare hybrid nanostructures with many advantages due to each kind of NM or NS (Bouakaz et al., 2015; Oraon et al., 2015; Alsharaeh et al., 2016; Chen et al., 2016; He 2016; Pérez del Pino et al., 2016,Kavinkumar et al., 2016; Zhao et al., 2016).These kinds of hybrid nanostructures with exceptional properties are effective and they were reported for the potential applications(Latorre-Sanchez et al., 2012;Lonkar et al., 2015).Many hybrid materialswere reported, which can bemade usingdifferent NS.But recently organic-inorganic hybrid NS haveshown considerable attention(Mishra et al., 2010; Chatterjee et al., 2013). Among the various organic NS materials, graphene oxide (GO)has a tremendous interest in a scientific research.Because GO istwo dimensional (2D) andconductinglayered NMand it consists of one atom-thick planar sheets of sp^2-bonded honeycomb structure of carbon atoms (Low et al., 2015).GO possesses extraordinary mechanical, thermal, electronic and physical properties,(Hansora et al., 2015;Lonkar et al., 2015),more importantly it contains a range of reactiveoxygen

functional groups (e.g., hydroxyl groups) (Yan et al., 2016).Therefore, GO wasused to prepareorganic-inorganic hybrid NS(Wu et al., 2012; Nandi et al., 2013; Esmaeili et al., 2014;Yadav et al., 2014; Angelopoulou et al., 2015, Low et al., 2015; Jain et al.,2016).Many scientists havepreparedhybrid NS usingdifferent metal and inorganic NM filled with GO, e.g., Titanium oxide–graphene(Heet al., 2016; Pérez del Pino et al.,2016; Zhao et al.,2016),Manganese-nickel mixed oxide/graphene (Latorre Sanchez et al.,2015), Organomontmorillonite/graphene(Bouakaz et al. 2015; Oraon et al., 2015),Calciumcarbonate/GO(Zhou et al., 2016),Silver nanoparticle/GO(Kavinkumaret al., 2016),Cobalt oxide nanoparticles/reduced GO(Alsharaeh et al.,2016),Zinc oxide nanorods/graphene(Chen et al., 2016),Vanadium dioxide nano flowers decorated on GO(Kang et al., 2016) and novel boehmite/GO nano-hybrids (Zhanga et al., 2016). Graphene based polymer hybrid nanocomposites werealsoreported (Acharya et al., 2007; Das and Prusty et al., 2013; Nandi et al., 2013; Chakraborty et al., 2014; Yadav et al., 2014; Angelopoulou et al., 2015; Bouakaz et al. 2015; Oran et al., 2015; Yana et al., 2016). But recently in the field of inorganic NM, layered double hydroxide (LDH) has emerged as the most powerful NMfor the preparation of multifunctional hybrids for various applications(Yuan et al., 2012). The LDH, a family member of inorganic layered NM, have recently attracted considerable attention because of their wide applications(Olfs et al., 2009; Ladewig et al., 2010; Chakraborty et al., 2012; Yuan et al., 2012; Lonkar et al., 2013; Menezes et al., 2014; Yang et al., 2014; Zazoua et al., 2014).The chemical composition of LDH is generallydescribed by the formula: $[M^{2+}_{1-x}M^{3+}_{x}(OH)_2]^{x+}\,[A^{n-}]_{x/n}\cdot mH_2O$. Usually, M^{2+} is a divalent metal (Ca^{2+}, Mg^{2+}, Zn^{2+}, Ni^{2+}, Co^{2+}, Mn^{2+}, Co^{2+} or Fe^{2+}), M^{3+} is a trivalent metal (Al^{3+}, Cr^{3+}, Mn^{3+}, Fe^{3+}, Co^{3+} or Ni^{3+}) and A^{n-} is a *n*-valent anion group (e.g. CO^{2-}_3, NO_3^-, PO_4^{3-}, SO_4^{2-}or Cl^-). The M^{2+}/M^{3+} ratio, also known as χ,usually lies in the range of $0.1 \leq \chi \leq 0.5$. LDH afford

space between the two layers and becomes suitable host for intercalation of planar transition metal complex catalyst containing porphyrin, phthalocyanine or Schiff base ligands (Sun et al.,2015). The LDH have a high anion exchange capacity and large surface area due to their structural configuration (Olfs et al., 2009;Basu et al., 2014). The nanostructures of these minerals arebrucite-like positively charged layers resulting from the partial substitution of original Mg^{2+} by Al^{3+}ions. The LDH can absorb inorganic as well as organic anions, which make them attractive materials for technological applications in different areas(Olfs et. al., 2009).Recently, graphene decorated layered metal hydroxides likeLDH(with different metal) compositions were studied in order to diminish their stacking interactions and to limit the aggregation ingraphenenanosheets(Garcia-Gallastegui et al. 2012; Latorre-Sanchez et al., 2012; Tang et al., 2012; Wen et al., 2013; Lonkar et al., 2015).In the present study, we report a simple method for preparation of multifunctional hybrid NS of Mg-Al-LDH@GO by bath ultra-sonication technique.Multi-layeredhybrids of LDH@GO were analyzed by Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), Field-emission scanning electron microscopy (FE-SEM), Transmission electron microscopy (TEM), Elemental analysis (EDX) and thermo gravimetric analysis (TGA)to investigate their functional composition, crystalline nature, layered surface morphology, elemental analysis and thermal behavior.

2. Experimental

2.1 Materials

For the synthesis of Mg-Al–LDH, magnesium nitrate ($Mg(NO_3)_2.6H_2O$) and aluminium nitrate ($Al(NO_3)_3.9H_2O$) (98% purity)werepurchased from Rankem, India. Sodium carbonate (anhydrous), Sodium hydroxide and sodium dodecyl sulphate (SDS)

were also purchased from Rankem, India and Sigma Aldrich,India respectively.For the synthesis of graphene oxide (GO),fine powder of graphite (98% purity) was purchased from LobaChem, India. Hydrogen peroxide (H_2O_2), Sulfuric acid (H_2SO_4) hydrochloric acid (HCl), sodium nitrate ($NaNO_3$)were purchased from Merck, India, while potassium permanganate ($KMnO_4$)was purchased from Rankem, India. All the chemicals were of analytical reagent grade and used without any further purification.Double distilled water was used throughout the experiments.

2.2 Synthesis of LDH@GOhybrid nanostructures

LDH@GObased hybrid NSwereeasily prepared by one-pot microwave-assisted synthesis approach (Garcia-Gallastegui et al. 2012; Tang et al., 2012; Wen et al., 2013; Lonkar et al., 2015). GO was prepared from graphite average particle size of < 20 μmby a modified Hummers method(Yadav et. al., 2014; Jain et al., 2016) and another simple and fast route is microwave assisted approach (Yan et al., 2016). Ultrasonic bath (Bio-Techniques, India) was usedto exfoliate GO sheets to make them individual sheets in an aqueous dispersion of water(Bourlinos, et al., 2009; Esmaeili, et al., 2014).Magnesium and aluminum (Mg-Al) based LDH were synthesized using $Mg(NO_3)_2{\cdot}6H_2O$ and $Al(NO_3)_3{\cdot}9H_2O$ by standard co-precipitation and thermal crystallization method (Acharya et al., 2007). SDS was used as a surface modifying agent forfunctionalization of the LDH, because SDS contains negatively charged functionality and highly nucleophilic sites in their chemical structure of LDH.Hence it is easy to make hybridsusing GO and LDH.The hybrids layered NS of LDH@GO were synthesized using bath ultrasonication technique.Same amount of LDH and GO (to keep mass ratio of 1:1) was taken in two different beakers and 50 mL of water was added in each beaker. The resultingdispersionswere sonicated for 15 min, and then LDH solution was added drop wise to the GO solution during bath sonication. The

resulting mixture was sonicated foradditional30 minwhich resulted information of a colloidal dispersion. The resulting dispersion of LDH@GO was allowed tosettle down for 48 h.Finally, the resultingproduct was vacuum dried at 120°C,which removed anyremaining traces of water.

2.3 Characterizations

The functional composition, crystalline nature, thermal behavior, surface and layered morphology, elemental composition and thermal behavior of GO sheets, LDH sheets and hybrid layered NS of LDH@GOhybridswere analyzed. The crystallinity was determined by X-ray diffractometer (XRD, Bruker D8 Advance, Berlin, Germany) in the range of $0\text{-}80^0$. The samples were placed vertically in front of the X-ray source. The detector was moving at an angle of 2θ, while the sample was moving at an angle of θ and the wavelength λ was 1.54 Å (Cu Kα, a tube voltage 40 kV and tube current 25 mA).

Functional groups of GO, LDH, and hybrid layered NS of LDH@GOwere analyzed by FTIR spectrophotometer (FTIR-8000 Spectrophotometer, Shimadzu, Tokyo, Japan). The number of scans per sample was kept 25 and resolution of the measurements was kept at 4 cm^{-1}. The recorded wave number range was keptin the range of 500-4000 cm^{-1}.

Surface morphology and EDS mappingofGO, LDH and hybrid layered NS of LDH@GO were determined by using field emission scanning electron microscope (FE-SEM, S-4800 Hitachi, Tokyo, Japan) operated at an accelerating voltage of 30 kV and transmission electron microscope (TEM, Philips CM-200, Eindhoven, The Netherlands) with 75 µA of filament current and 200 kV of accelerating voltage.For analyzing elemental composition of LDH@GO, EDSspectra were recorded on spectrometer attachedwith the FE-SEM.

Thermal stability of GO, LDH, and hybrid layered NS of LDH@GOwas determined by a thermo gravimetric analyzer (TGA-50, Shimadzu, Tokyo, Japan). Powder form of all samples wasusedfor thermal analysis.TGA wasrecorded at room temperature. Approximately10 mg ofsample was placed in a platinum pan for TGA analysis. The temperature range was kept from 30to 700°C and a heating rate was kept 10°C/min under nitrogen atmosphere to avoid thermoxidative degradation.

3. Results and discussion

3.1 Structural and crystallinity analysis

The crystal structure as well as orientation were studied from XRD analysis which also veried the average spacing between GO, LDH,hybrid layered NS of LDH@GO.Fig. 1andFig. S4show the XRD patterns of GO, uncalcined LDH, calcined LDH, modified LDH andhybrid layered NS of LDH@GO.FromFig. 1 (a),it can be seenthatthe diffraction peak of exfoliated GO was recorded at $2\theta = 10.11^0$, whichattributes to the plane of GO (002)features basal spacing of 8.73 Å. This also shows the completeoxidation of graphite into the GO due to the introduction ofoxygen-containing functional groups on the graphene sheets(Lonkar et al., 2015).The characteristic peaks are appeared as sharp in the XRD patterns of uncalcined LDH (Fig. S4) the peaks at 2θ =11.7, 23.5^0, 34.9^0, 39.5^0, 47.1^0, 60.9^0 and 62.4^0 which are corresponding to the (003) (006) (012) (015) (018) (110) (113) planes.The XRD pattern of calcined LDH (Fig. 1b) shows weak reflections, confirms the destruction of layered structure upon calcinations and formation of metals oxides. From XRD patternof(Fig.1(c),anintercalation of SDS anionscould be seen after modification with SDS due to virtue of an increase in basal spacing. Hence peak shiftedslightly towards lower diffraction angle(Chakraborty et al., 2014). This also indicatesthe expansion in the interlayer distance and these planes also

show the characteristic peaks of Mg/Al based LDH(Lonkar et al., 2013; Yang et al.,2014; Zazoua et al.,2014).The XRD patterns(Fig. 1d)of the as-prepared LDH@GO hybrid NSshowamorphous nature,but nocharacteristic peak of GO was observedin hybrid layered NS ofLDH@GO.The peaksat34.0^0 and 60.0^0, at (012) and (110) intensitiescorrespond to LDH. The peak of GO at 2θ = 10.11^0(002), shown in Fig.1(a),was significantly broadened and shifted towardslower diffraction anglebecause of increased distance between GO sheets due to incorporation of exfoliated LDH(Latorre-Sanchez et al., 2012;Lonkar et al., 2015).

3.2 Functional groupanalysis

The FT-IR spectra of GO, calcined LDH, modifiedLDH and hybrid layered NS ofLDH@GOare shown in Fig. 2(a-d).The strongand broad peak at 3360 cm^{-1}seen in Fig. 2(a) indicates the presence of surface O–H stretchingdue to vibrations of the H-O-H groups of water. The other peakscorresponds to oxygen functional groups, such as carboxyl(C=O) stretching of COOH groups (1725 cm^{-1}), aromatic(C=C)stretching (1615 cm^{-1}), epoxy (C–O) groupstretching (1218 cm^{-1}) and alkoxy(C–OH) group stretchingvibrations (1043 cm^{-1})(Wen et al., 2013;Esmaeili et al., 2014; Low et al., 2015).The FTIR spectrum of the calcined LDH (Fig. 2b)displaythe broad and strong bands in the range of 3200-3600cm^{-1} and the other low frequency region (800 cm^{-1}) is attributed to metal–oxygen and metal–hydroxyl vibration modes present in the lattice of LDH.The FTIR spectrum of the modified LDH (Fig. 2c)shows two types of bands; firstis corresponding to the anionic species of SDS intercalated and second is corresponding to the pristine LDH. It also shows the stretching band for aliphatic CH_3 of the long chain of SDS molecules around 2854-2965 cm^{-1}. The bands at 1216 and 1063 cm^{-1} are ascribed to the symmetric vibration of sulfate from SDS, respectively (Chakraborty et al., 2014; Jain et al., 2016).The other low frequency regions(800 cm^{-1})

are attributed to metal–oxygen and metal–hydroxyl vibration modes present in the lattice of LDH. (Yang et al.,2014).The broad band in the range of 3200–3700 cm^{-1}may be due to presence of O-H stretching vibration of hydrogen bonded metal hydroxide layer and interlayer water molecules(Lonkar et al., 2013; Chakraborty et al., 2014; Zazoua et al.,2014). The band observed near 1637 $cm^{-1}$is assigned to the bending vibration of water molecules. From the peak intensitiescorresponding to GO and LDHshown inFig. 2(d), it can be said that carbonyl, epoxide, and ether groups wereobserved to be weakenedin the FTIR spectrum of LDH@GOhybrids. This also confirms the formation of hybrid nanostructures.Moreover, the XRD patterns(Fig.1 d)and TEMimages(Fig. 4 c and Fig. 4d) confirm the inter layer exfoliated hybridization.

3.3 Surface morphology and elemental analysis

Typical FE-SEM and TEM images of GO, calcined LDH, modified LDH and LDH@GO hybrid NSare shown in Fig. 3 and Fig. 4. GO exhibited the exfoliated layers structure with scrolled multilayer sheets with thin, transparent, smooth, wrinkles and folding on the basal and edges with an average 50 nm in thickness and up to the certainμm in length (Fig.3a and Fig.4a)(Oraon et al., 2015; Jain et al., 2016; Kavinkumaret al., 2016, Pérez del Pino et al., 2016;Yan et al., 2016;).The GO sheets are generally exfoliated due to ultrasonic vibrations and LDH are get intercalated in between GO sheets.As shown in Fig. 3(c)and Fig. 4(b), the LDH consist of regular and thin hexagonal single platelets with exfoliated structures with anaverage thickness of 25 nm and length up to the 250nm, also indicatessize and morphology at nanoscale(Chakraborty et al., 2012; Diao et al., 2014)as compared to calcined LDHshown in Fig. 3(b) and also TEM images of LDH are inserted in Fig.4(b).

The surface morphology of the hybrid NS of LDH@GOwas also studiedby FE-SEM, TEM (Fig. 3(d), and Fig. 4(c-d). The corresponding images show significant

morphological differences with respect to the GO. Based onmicroscopicand XRD (Fig. 1c)analysisof hybrid NS of LDH@GO,it can be consideredthatLDH has been uniformly inserted into GO sheets and considerately amplifies the distances between adjacent GO sheets (Scheme 1) (Zhao et al., 2014, Lonkar et al., 2015; Kang et al. 2016;Kavinkumaret al., 2016; Zhang et al.,2016).According to morphological results, a schematic representationofthe formation of LDH@GO hybrid NS materialis shown in Scheme 1. LDH is positively charged ions and GO with a basal spacing at nanoscale could be considered as a negatively chargedsingle sheet of graphene due to the presence of carboxylate. The LDH nanoplatelets are adsorbed on the surface of GO nanosheetsdue to the electrostatic interaction between GO and LDH.The intercalation of LDH into the nanosheetsof graphene can effectively prevent the restacking of GO nanosheets (Zhao et al., 2014).

Formation of hybrid NS ofLDH@GOwas further confirmed by EDS analysis, which showed presence of these different elements such as C, O, Mg and Al peaks as well as their relative quantities (Fig. 3e).The EDS spectra as well as elemental mapping of GO, LDH and LDH@GO further confirms the presence of LDH incorporated in GO sheets (Fig.S1, Fig. S2 and Fig. S3given insupplementary information).

3.4 Thermal behavior

The TGA behaviors of the GO, LDH and hybrid NSof LDH@GOaredisplayed in Fig. 5. The pristine GO exhibits single stepdegradation with significant mass loss at around 180°C due to the pyrolysis of the labile oxygen-containingfunctional groups.The presence of the oxygen functional groups makes GO thermally unstable, as itundergoes pyrolysis at elevated temperatures (Fig. 5a), mass loss was recorded up to 90% below 200°C mainly due to thedecomposition of oxygen-containing groups and the loss of interlayer water molecules(Esmaeili et al., 2014).The modified LDH show typical two

stage thermal decompositionpattern with 45% mass loss (Fig. 5b). The consecutive loss of waterpresent in the LDHwas observed from room temperatureto 200°C, chemisorbed at 200–360°C and the water arisingfrom the dehydroxilation of the layers at 225–450°C. The TGA pattern of hybrid NS ofLDH@GOwith 67% mass loss (Fig. 5c), shows three major mass loss stages. The first massloss observed at approximately 210°C, isattributed to the removal of loosely bound water moleculesfrom the LDH interlayer. The secondmassloss, in thetemperature range 250–350°C, is due to the removal of oxygen functionalities. The third and final masslosswas observed above temperature of350°C, which is primarily dueto dehydroxylation and decarbonation of the LDH sheets(Lonkar et al., 2015)and thus the TGA graph showed 22 % GO hybrid with LDH.

4. Conclusion

Hybrid layered NSof LDH@GOwere synthesized by an efficient and rapid bath ultra-sonic technique. The resulting hybrid layered NS of LDH@GOshowedimprovement inspacing between the GO sheets Moreover, the hybrid layered NS of LDH@GOdemonstrated good thermal stability as comparedtopristine GO. Hence, conjunction of GO and LDH hybrid NSendow the multifunctional properties in a single hybrid system. The effectiveness of themethod described thesynthesis ofLDH@GO hybrid layered NS and can be useful for the preparation of other graphene-based hybrid NS. Similarly, the use of bath ultra-sonicationprovidedproper exfoliation of GO sheets for thedevelopment of multi-functional hybrid layeredNS.Anincreased distance between GO sheets and improved thermal stability suggest that LDH nanoplatelets may be incorporated in between GO sheets during bath sonication. Further studies on the relationship between the solution reflux casting of hybrid layered NS ofLDH@GOand

theirintercalation and exfoliationmechanismin polymer matrices along with their detailed investigations of the physico-mechanical(multifunctional) propertiesare future prospective of this work.

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Captions for Figures

Fig. 1. XRD diffraction pattern of (a) GO (b) Calcined Mg-Al LDH (c) Modified Mg-Al LDH (d) Hybrid layeredNS of Mg-Al-LDH@GO.

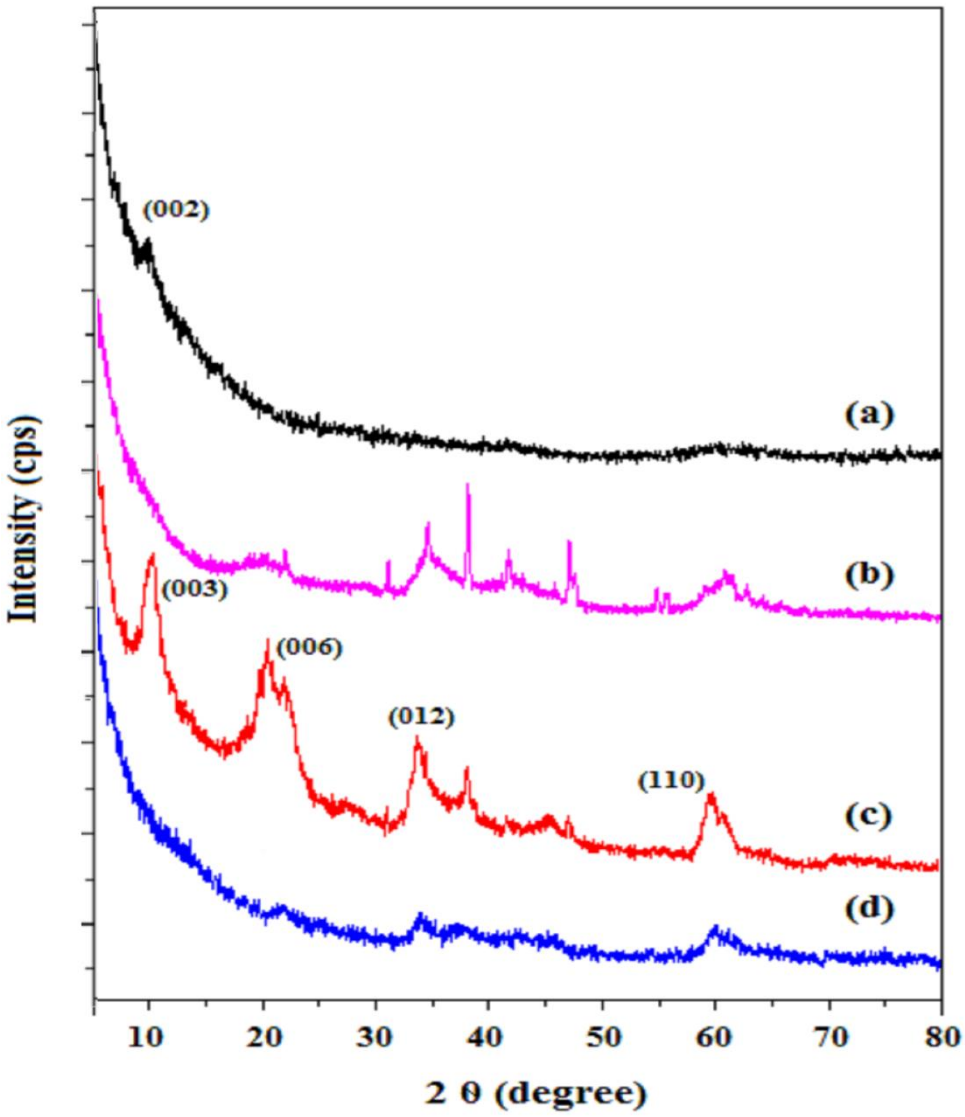
Fig. 2 FTIR spectra of (a) GO (b)Calcined Mg-Al LDH (c) ModifiedMg-Al LDH (d) Hybrid layeredNSof Mg-Al-LDH@GO.

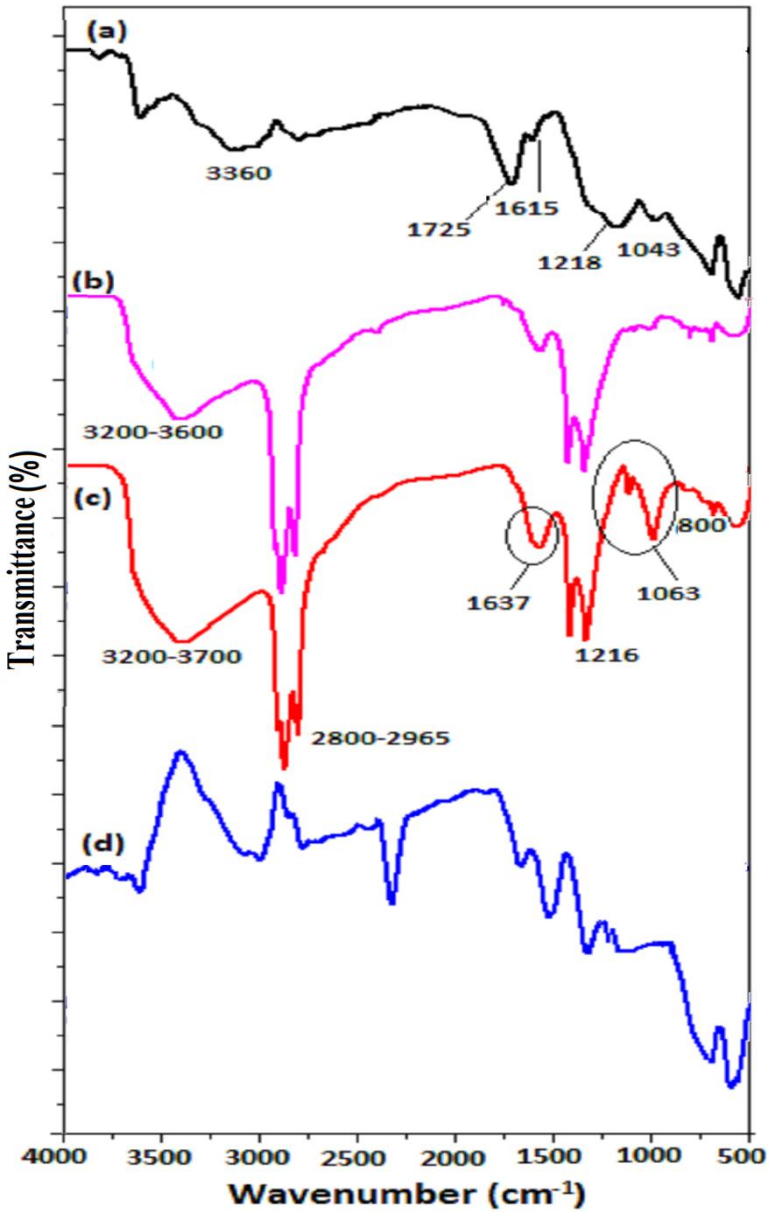
Fig. 3. FE-SEM images of, (a) GO (b) Calcined Mg-Al LDH (c) Modified Mg-Al LDH (d) Hybrid layeredNS of Mg-Al-LDH@GO (e) Elemental analysis of Mg-Al-LDH/@GO based nanostructures.

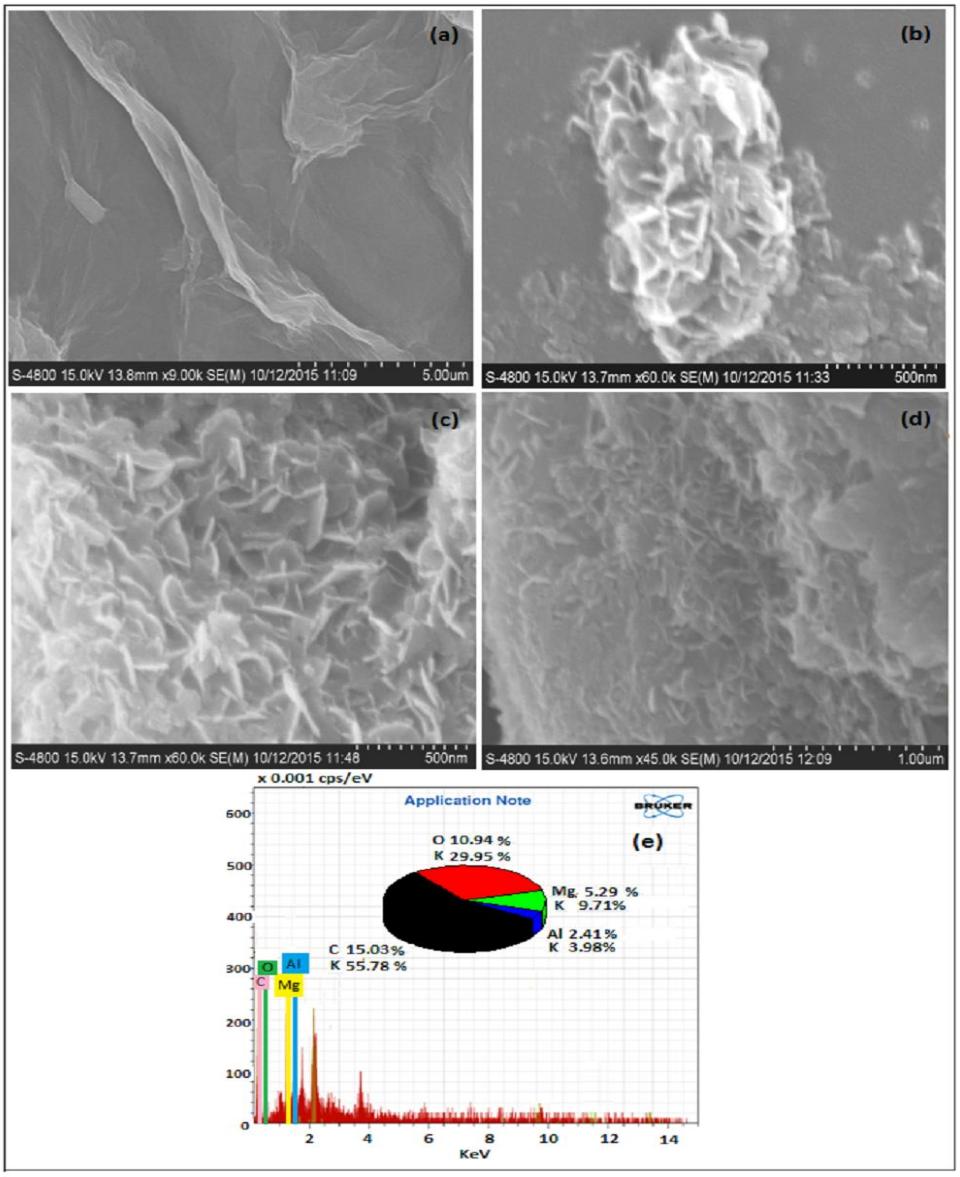
Fig. 4. TEM images of (a) GO (b) Mg-Al LDH (c-d) Hybrid layeredNS of Mg-Al-LDH@GO.

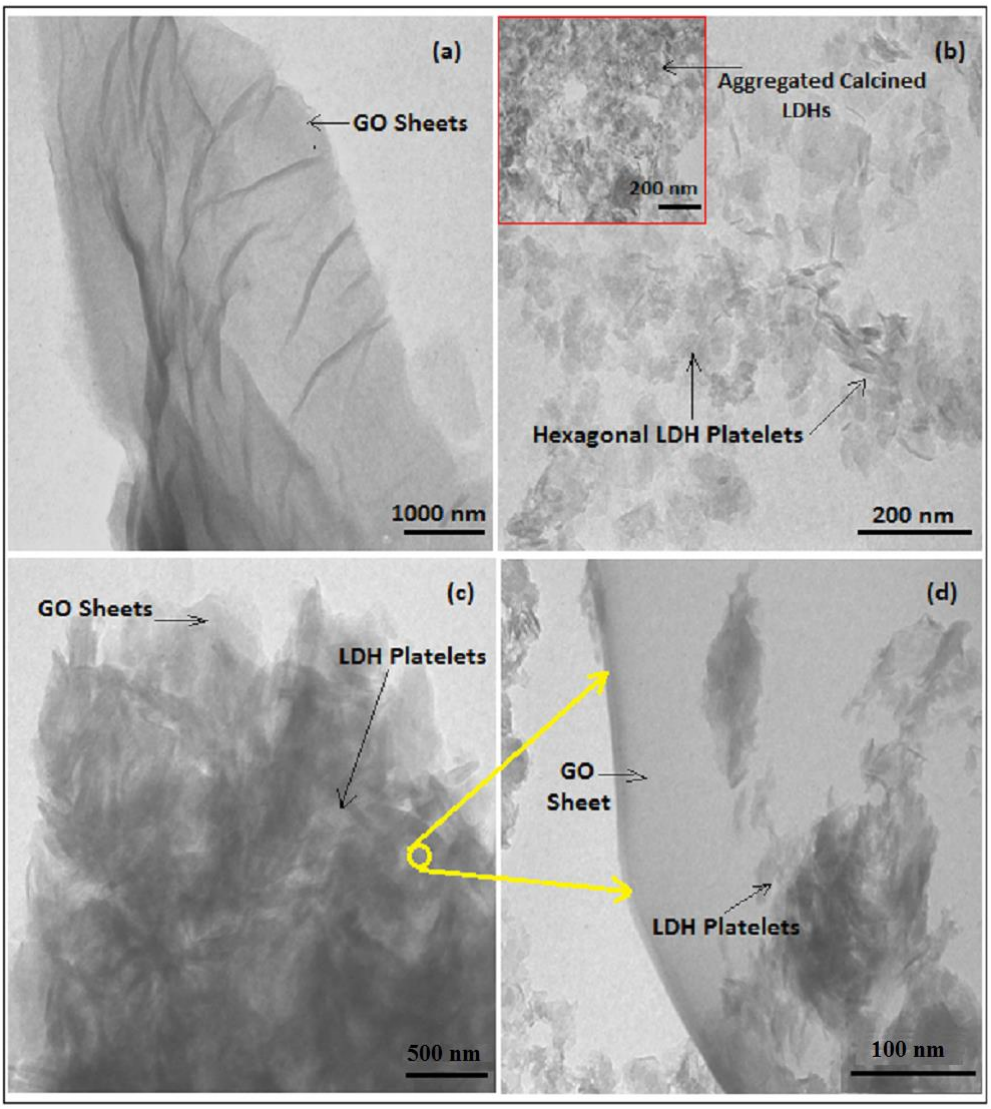
Fig. 5. TGA curves of (a) GO (b) modified Mg-Al LDH (c) Mg-Al-LDH@GObased hybrid nanostructures.

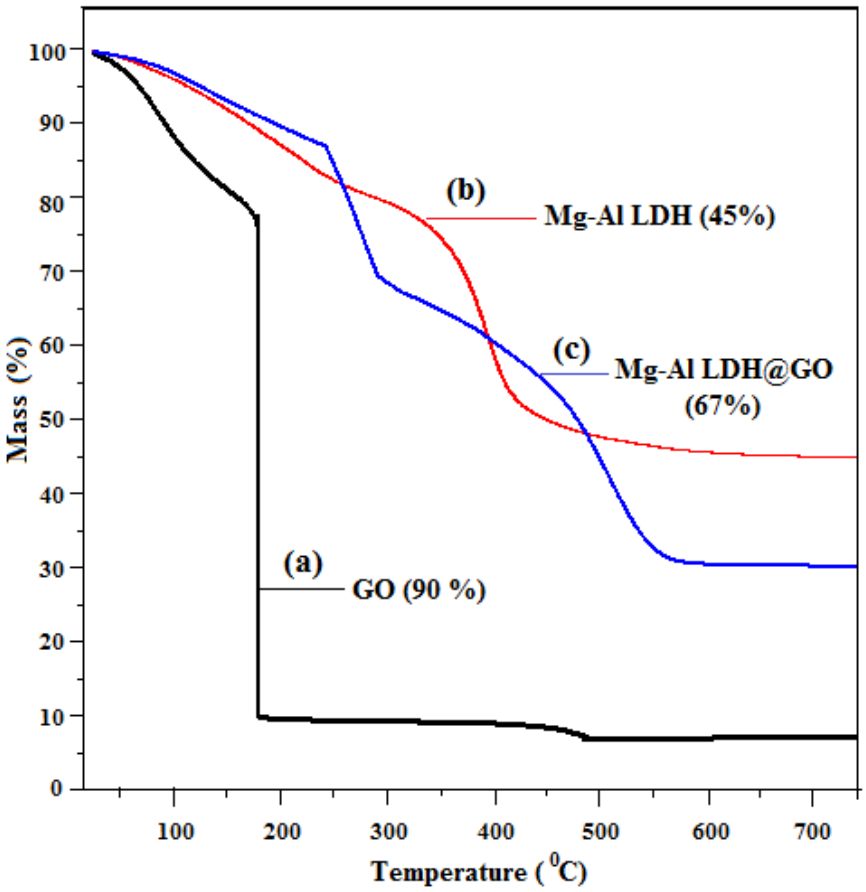
Scheme 1. Pictorial representation of the formation of interlayer hybrid layered nanostructures of Mg-Al-LDH/@GO prepared by bath ultrasonication technique.

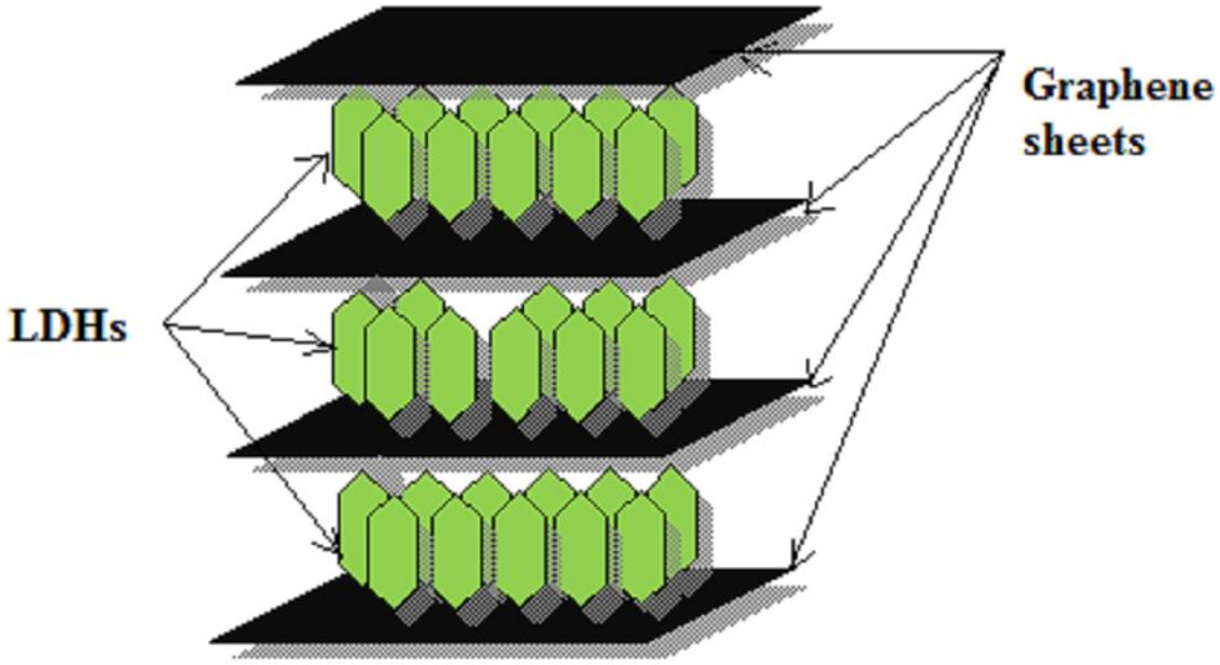

Intensity (cps)
(002)
(a)
(b)
(003)
(006)
(012)
(110)
(c)
(d)
10
20
30
40
50
60
70
80
2 θ (degree)


(a)
3360
1725
1615
1218
1043
(b)
3200-3600
(c)
3200-3700
1637
1216
1063
800
2800-2965
(d)
Transmittance (%)
4000
3500
3000
2500
2000
1500
1000
500
Wavenumber (cm-1)


(a)
S-4800 15.0kV 13.8mm x9.00k SE(M) 10/12/2015 11:09 5.00um
(b)
S-4800 15.0kV 13.7mm x60.0k SE(M) 10/12/2015 11:33 500nm
(c)
S-4800 15.0kV 13.7mm x60.0k SE(M) 10/12/2015 11:48 500nm
(d)
S-4800 15.0kV 13.6mm x45.0k SE(M) 10/12/2015 12:09 1.00um
x 0.001 cps/eV
Application Note
BRUKER
(e)
O 10.94 %
K 29.95 %
Mg 5.29 %
K 9.71%
Al 2.41 %
K 3.98%
C 15.03%
K 55.78 %
O
Al
C
Mg
600
500
400
300
200
100
0
2
4
6
8
10
12
14
KeV


(a)
GO Sheets
1000 nm
(b)
Aggregated Calcined LDHs
200 nm
Hexagonal LDH Platelets
200 nm
GO Sheets
(c)
LDH Platelets
500 nm
(d)
GO Sheet
LDH Platelets
100 nm


Mass (%)
Temperature (^{0}C)
(a)
GO (90 %)
(b)
Mg-Al LDH (45%)
(c)
Mg-Al LDH@GO
(67%)



Intercalation of LDHs in between exfoliated graphene sheets

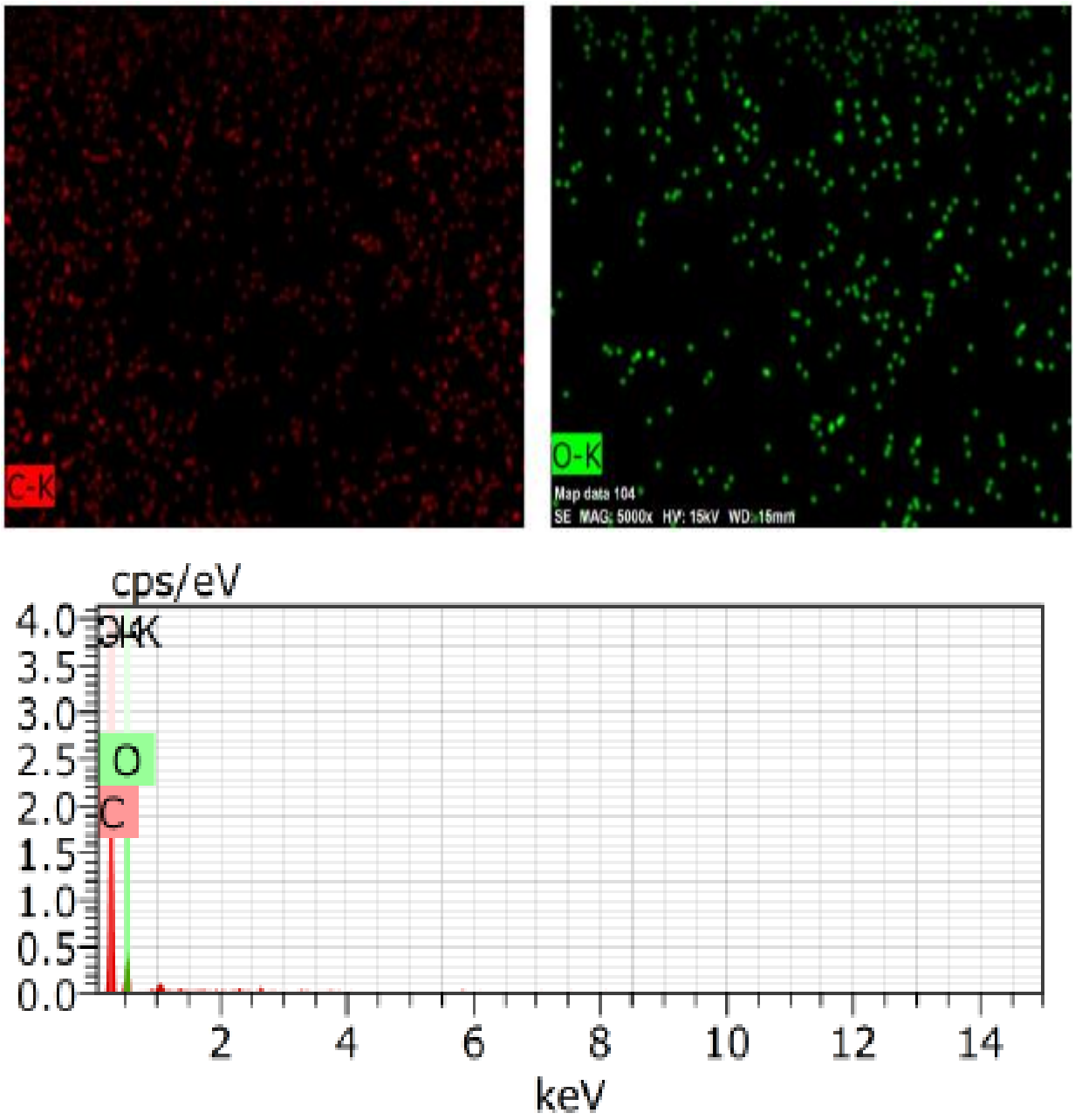


Fig. S1.FE-SEM EDS and Elemental mapping spectrum analysis indicating corresponding elementalmapping C and Oin GO.

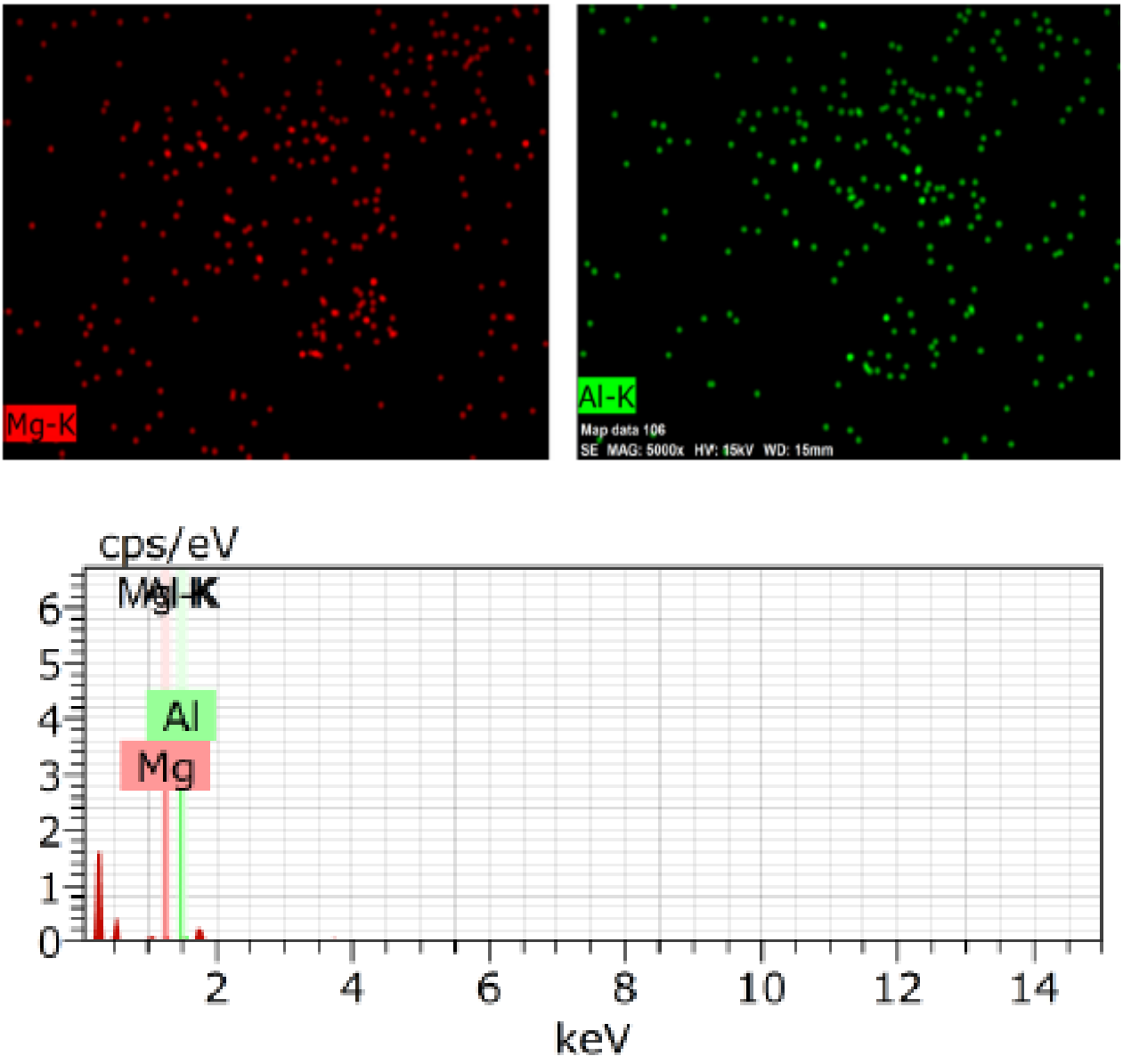


Fig. S2. FE-SEM EDSand Elemental mapping spectrum analysis indicating corresponding elementalmapping of Mg and Al in Mg-Al LDH.

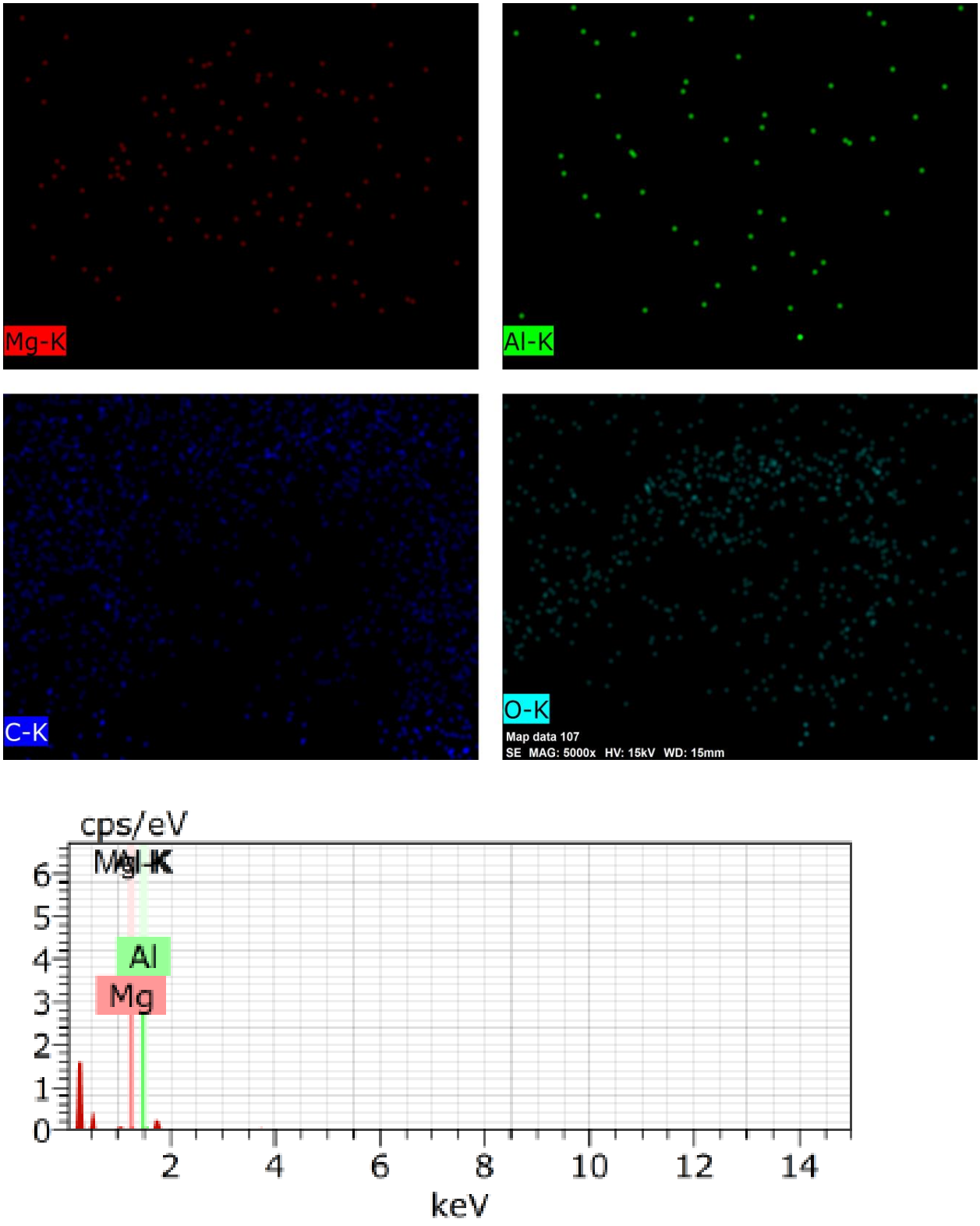


Fig. S3.FE-SEM EDSand Elemental mapping spectrum analysis indicating corresponding elementalmapping C, O, Mg and Al inhybrid NS of LDH@GO.

FE-SEM EDS pattern and elemental mapping of GO, LDH and hybrid NSs of LDH@GO was shown in Fig. S1, S2 and Fig. S3.The carbon and oxygen distribution in the plane of graphene indicates highly homogeneous, as can be seen by the elementalmapping shown in Fig. S1. The same C, and O mapping of graphene suggests that not only the edge but also the plane ofvgraphene contain a large amount of C and O functionalities. Similarly, magnesium and aluminum distribution in Mg-Al-LDH can be seen from the by the elementalmapping shown in Fig. S2, which indicates that LDH contain sufficient amount of Mg and Al. The EDX spectra as well as elemental mapping of and LDH@GO further confirms the presence of LDHs incorporated in GO sheets C and Ofor GO, Mg and Al for LDH and C, O, Mg and Al for LDH@GOhybrid nanostructures, respectively (Fig. S3). Theimages (Fig. S3)also suggestthat the LDH are homogenously inserted/ distribution in GO sheets. And EDX pattern describes the presence of LDH in GO sheets.

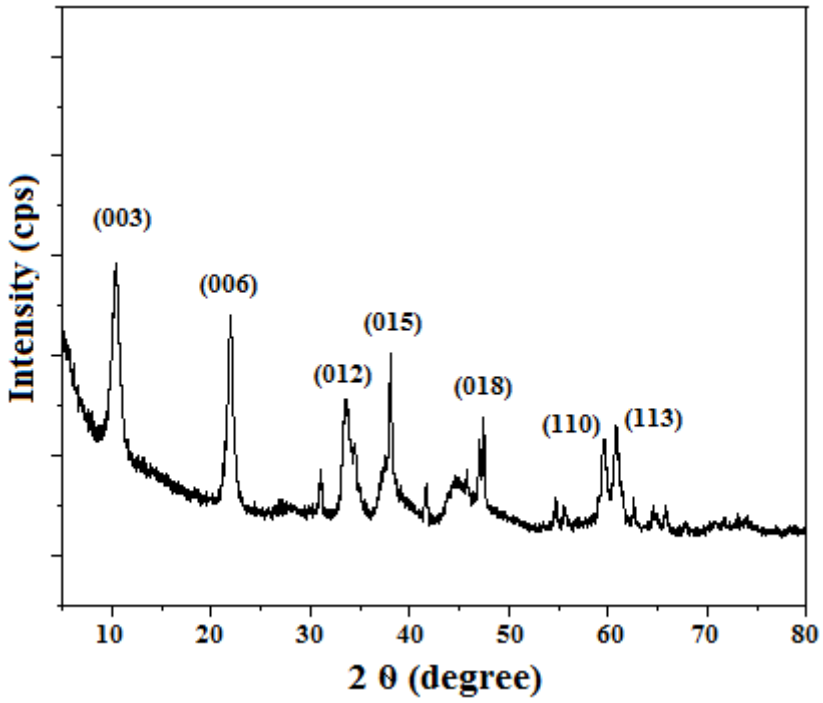


Fig. S4.XRD diffraction pattern of uncalcined Mg-Al LDH

The characteristic peaks are appeared as sharp in the XRD patternsof uncalcined LDH (Fig. S4) the peaks at 2θ =11.7, 23.5, 34.9, 39.5, 47.1, 60.9 and 62.4^0which are corresponding to the (003) (006) (012) (015) (018) (110) (113) planesand these planes also show the characteristic peaks of Mg-Al-LDH