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Carbon-layered double hydroxide nanocomposite for efficient removal of inorganic and organic based water contaminants – unravelling the adsorption mechanism

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Carbon-layered double hydroxide (C-LDH) nanocomposites were synthesized by a simple homogeneous co-precipitation process using as-prepared porous carbon and Al, Mg, Ca precursor salts of LDH. The synthesized adsorbent was used for the removal of inorganic (fluoride, arsenic, iron) and organic (cationic dye methylene blue, MB and anionic dye methyl orange, MO) based water contaminants separately and simultaneously from water. Microstructural analysis revealed the formation of nano-flake-like LDH particles (dia = 10-15 nm and length = 50-100 nm) adhered onto the surface of carbon nanospheres (50-150 nm). The surface area of the products was in the range of 758-477 m²/g. It rendered maximum adsorption capacities of 22.37, 20.40, 80, 122.1 and 328.95 mg/g for As(V), F⁻, Fe(II)/Fe(III), MB and MO, respectively. It was used for simultaneous removal of multi-contaminants like As(V), F⁻, Fe(II)/Fe(III), MB and MO from water with % adsorption up to 99.99%. This study illustrates a synergetic effect of composition (C : LDH mol ratio) and surface properties (total surface area and mesopore : micropore surface area ratio) of the adsorbent on their relative adsorptions of various pollutants.

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Introduction

Water pollution by organic and inorganic contaminants has drawn global concern in recent years. Various metal ions and organic toxic pollutants are discharged into environment which is very harmful to human beings and aquatic life, leading to severe damage for the whole ecosystems. Over the last few decades, the level of water pollution has increased worldwide at an alarming rate due to rapid industrialization. Release of several inorganic (arsenic, fluoride, iron, lead, and cadmium etc.) and organic (phenol, different dyes) pollutants from different industries like pharmaceutical, leather, power plants, pesticide, textile, coal, mines, petrochemical, heavy metal industries etc., and also from weathering of rocks has contaminated water bodies. The presence of arsenic and fluoride, and organic dyes like methylene blue and methyl orange in water causes serious hazardous effect on living beings.

Numerous approaches have been developed for removal of water contaminants like ion exchange, coprecipitation, electrocoagulation, membrane separation, adsorption, photocatalysis etc. However, most methods have limitation due to high investment cost, secondary pollution and poor efficacy. Accordingly, these methods are restricted towards the application in waste water treatment. However, adsorption is considered as most efficient and promising method due to its large adaptability in small scale treatment plants, low cost, simple design, regeneration ability and ease of operation. Several adsorbents including alumina, activated carbon, clay, zeolite etc. have been widely used for removal of water contaminants. Nanomaterials are found to be effective adsorbents for water decontamination due to their high surface to volume ratio. A wide number of studies have reported the removal of water contaminants using carbon-based nano-adsorbents containing high surface area and presence of abundant surface functional groups. For the removal of water contaminants specifically organic pollutants, carbon-based nano-adsorbents are used because of their high surface area. However, due to the presence of high microporosity (< 2
nm pore size) and absence of any ionic counterparts in carbon. Therefore, carbon based adsorbents have some limitation for the adsorption of most of the inorganic water pollutants via electrostatic interaction. In recent days, inorganic layered double hydroxide (LDH) based materials have gained integrative attention due to their distinctive physicochemical properties.\(^{19}\) Generally LDH are represented as \([\text{M}^{2+}(1-x)\text{M}^{3+}x(\text{OH})_2]^{x+}(\text{A}^{n-})_{x/n}\cdot y\text{H}_2\text{O},\) where the \(\text{M}^{2+}, \text{M}^{3+}\) and \(\text{A}^{n-}\) represent the divalent (Co\(^{2+}\), Cu\(^{2+}\), Ca\(^{2+}\), Mg\(^{2+}\), Zn\(^{2+}\), Ni\(^{2+}\), Mn\(^{2+}\)) metal cations, trivalent metal cations (Al\(^{3+}\), Mn\(^{3+}\), Co\(^{3+}\), Fe\(^{3+}\), Cr\(^{3+}\)) and the intercalating anions, respectively. LDH based adsorbent showed effective adsorption efficiency towards the removal of anionic contaminants or cationic pollutants via ion exchange with \(\text{M}^{2+}\) ions in LDH due to the presence of large interlayer space with exchangeable anions and positively charged layer.\(^{20,21}\) However, LDH based materials due to their lower surface area than carbon based adsorbents have some restriction for the removal of organic water pollutants. It is worth mentioning that the multiple oxygenic functional groups on the surface of carbon could help facilitate in the formation of carbon based composites in the presence of other adsorbent material like LDH. Therefore, carbon based LDH composite (C-LDH) could be effective for the removal of water contaminants via physical adsorption of metal ions on the carbon surface as well as exchange of metal ions with the interlayer anions of LDH through the formation of chemical bonding with surface hydroxyl group or other oxygen groups on the carbon surface.\(^{22,23}\) The synergistic effect of both the adsorbents i.e., porous carbon and layered double hydroxide in the form of composite could improve their adsorption capacity by removing different inorganic and organic water contaminants simultaneously. Huang \textit{et al.}\(^{24}\) stated the removal of Pb(II) and humic acid from aqueous solution using layered double hydroxide/hollow carbon microsphere composites. Fe\(_3\)O\(_4\)@C@Layered double hydroxide composite was used as a magnetic adsorbent for the removal of uranium.\(^{25}\)
With the above motivation, the objective of the present work is to synthesize an effective adsorbent i.e., carbon-layered double hydroxide (C-LDH) nanocomposite for simultaneous removal of inorganic (As(V), Fe(II)/Fe(III) and F⁻) and organic (methylene blue, MB and methyl orange, MO dyes) water contaminants. The efficiency of the nanocomposite materials has been investigated by varying different parameters like time, pollutant concentration, temperature and pH of the solution. In this study, a synergetic effect of composition (C : LDH mol ratio) and the surface properties (BET surface area and mesoporous : microporous surface area ratio) of the composite has been illustrated for the adsorption of different water contaminants. A mechanistic understanding of adsorption process for simultaneous removal of different water pollutants has been highlighted. The present work is significant in developing a unique adsorbent material for the removal of all possible water contaminants originated from industrial effluents as well as contaminated natural ground water to be used as domestic purpose.

Materials and methods

Materials

Aluminium nitrate nonahydrate (Al(NO₃)₃.9H₂O, assay > 99 %), calcium nitrate dihydrate (Ca(NO₃)₂.4H₂O, assay 99.0 - 102.0 %), magnesium nitrate hexahydrate (Mg(NO₃)₂.6H₂O, assay ≥ 98.5 %), ammonium hydroxide (NH₄OH, 25%), methylene blue, methyl orange, sodium fluoride (NaF, assay ≥ 97 %), ferrous sulphate (FeSO₄. 7H₂O, assay 99.5 - 102.0 %) and arsenic (1000 mg/L) standard solution were purchased from Merck, India, while sodium arsenate heptahydrate (Na₂HAsO₄.7H₂O, 98-102.0 %) obtained from Loba Chemie. Millipore water was used throughout the experiment.

Synthesis procedure

Carbon-layered double hydroxide (C-LDH) composite was synthesized by a simple homogeneous precipitation process. A solution of Al(NO₃)₂, Mg(NO₃)₂ and Ca(NO₃)₂ were
prepared with 70 mL Millipore water maintaining their molar ratio of 1 : 1.33 : 0.66, respectively (Solution A). As-prepared porous carbon (ESI†) was added into the solution A under stirring for 24 h maintaining different molar concentrations (0.015 to 0.06 mols) with respect to one unit of LDH (as prepared in solution A). 2 mL NH₄OH (25 wt %) in 20 mL water was added dropwise into it followed by warming at 60 ºC for 4 h under stirring (Solution B). After that, the products (C-LDH nanocomposite) were collected via washing with water and drying at 100 ºC. Thus, four different samples were prepared designated as C₄ₓ-LDH, C₂ₓ-LDH, C₃ₓ/2-LDH and Cₓ-LDH with carbon contents of 0.06, 0.03, 0.02 and 0.015 mols, respectively, where x is the amount of carbon (in mol) with respect to 1 Al³⁺ : 1.33 Mg²⁺ : 0.66 Ca²⁺ (mol ratio) of LDH.

Characterization

The synthesized products were characterized by XRD (PhilipsX’Pert Pro PW 3050/60, using Ni-filtered Cu-Kα radiation, λ= 0.15418 nm), FTIR (Spectrum two, PerkinElmer), N₂ adsorption-desorption study, XPS (ULVAC-PHI), FESEM (Model: Zeiss, Supra™ 35VP, Oberkochen, Germany) and TEM (Tecnai G2 30ST (FEI). In the study of N₂ adsorption-desorption, BET (Brunauer-Emmett-Teller) method was used to measure the specific surface area of the samples, while BJH (Barrett-Joyner-Halenda) method was adopted to evaluate the pore size distributions of the C-LDH samples. Aluminum, magnesium and carbon contents of the sample were analysed by Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES), ARCOS 130 MV, Spectro Analytical Instruments GmbH, Kleve, Germany), and carbon content was analysed by C-600 Carbon Analyser, Leco, USA.
To record UV–Visible spectra, UV-Vis spectrophotometer (Jasco V-730) was used within the wavelength range of 195 to 1100 nm.

Adsorption experiment

The experiments for the adsorption of arsenate, fluoride, iron, methyl orange and methylene blue on C-LDH composites were carried out by changing different experimental parameters like time, adsorbate & adsorbent concentration, pH and temperature (pollutant concentration, adsorbent dose, time, pH and temperature for each experiment are given in the respective section of Results and Discussion part). After specified time interval, adsorbents were separated from aqueous solution by filtration. The absorbance values were measured at adsorbent maxima of the respective pollutants. The concentration of pollutant in the filtrate was measured using UV-Vis spectrophotometer. The adsorption capacity (\(q_t\) in mg/g) and % removal of pollutants were estimated by the following equations.

\[
q_t = (C_o - C_t).V/m \quad (1)
\]

\[
\% \text{ of pollutants adsorbed} = \frac{(C_o - C_e)}{100 \ C_o} \quad (2)
\]

where, the concentrations of pollutant (mg/L) at initial time, at time \(t\) and at equilibrium time is defined by \(C_o\), \(C_t\) and \(C_e\), respectively. \(V\) and \(m\) are the volume (mL) of the solution and mass of the adsorbent (gm), respectively.

Results and discussion

Characterization of synthesized C-LDH composite

The XRD pattern of synthesized C-LDH composites is shown in Fig. 1a. The characteristic peaks of layered double hydroxides (LDH) with 20 values of 11.5, 23.4, 34.9, 39.6, 60.9 and 61.1 corresponding to hkl planes of (003), (006), (101), (012), (015), (110) and (113), respectively are identified.\(^{27}\) Figure S1, ESI† represents the plot of normalized intensity vs 20 (degree). By changing different molar compositions of C : LDH, there is no significant
changes in XRD peak position of the composites. However, the characteristic peak intensity of LDH increases with increase in content of LDH in the composites. FTIR spectra of C-LDH composites are shown in Fig. 1b. The absorption bands at 446 and 654 cm\(^{-1}\) correspond to the Al-O and Mg-O lattice vibrations whereas peak position at 1379 cm\(^{-1}\) is due to the presence of nitrate/carbonate anions in the composites. The wide absorption band at 3446 cm\(^{-1}\) and the sharp band at 1629 cm\(^{-1}\) are the characteristics stretching and bending vibrations of O-H bond, respectively.

![Fig. 1](image_url)

**Fig. 1** (a) XRD pattern and (b) FTIR spectra of C-LDH composites: C\(_{4x}\)-LDH, C\(_{2x}\)-LDH, C\(_{3x/2}\)-LDH and C\(_{x}\)-LDH.

Fig. 2 shows N\(_2\) adsorption-desorption isotherms of the four different C-LDH composites, insets reveal the BJH pore size distribution (desorption) of the respective samples. Isotherms reveal pseudo-type I and pseudo-type IV isotherms indicating the presence of microporosity and mesoporosity in the samples, respectively. It signifies that both the micropores and mesopores are present in each sample. Notably, for all the samples C\(_{4x}\)-LDH, C\(_{2x}\)-LDH, C\(_{3x/2}\)-LDH and C\(_{x}\)-LDH, there occurs a steep rise of isotherms at lower relative pressure (\(p/p_o < 0.01\)) and at higher relative pressure (\(p/p_o < 0.8\)); the former is due to the presence of microporosity while the latter demonstrates interparticle mesoporosity in the samples. Table 1 represents the BET surface area, pore volume and pore diameter of the samples. The porous carbon sample used for the preparation of C-LDH composite has BET surface area and pore volume of 752 m\(^2\)/g and 1.155 cc/g, respectively. During formation of
C-LDH composite, the molecular species of LDH precursors could block partly the interparticle mesopores of carbon particles, thereby decreasing the BET mesopore surface area and pore volume of the composite (Table 1). The pore diameter of the samples is in the range of 3.6-3.9 nm while the sample C_{4x}-LDH shows bimodal pore size distribution at around 1.2 nm and 3.6 nm (inset of Fig. 2a). It is to be noted that except for the sample C_{3x/2}-LDH there is a decrease trend of total BET surface area, micropore surface area and mesopore surface area in the order of C_{4x}-LDH > C_{2x}-LDH > C_{x}-LDH. However, the pore volumes for all the samples decrease in the order of C_{4x}-LDH > C_{2x}-LDH > C_{3x/2}-LDH > C_{x}-LDH. It is worth mentioning that with decrease of carbon content in the composite, the total BET surface area, micropore surface area, mesopore surface area and pore volume decreased. The anomalous result of the sample C_{3x/2}-LDH in terms of total surface area, mesopore and micropore surface area could be due to the shape of pore geometry. Interestingly, Fig. 2c shows that hysteresis loop of the sample C_{3x/2}-LDH is more expanded compared to other samples. It is worth mentioning that all the samples depicts H3 type hysteresis loop indicating slit like mesopores. For the sample C_{3x/2}-LDH, the more expanded hysteresis loop of the isotherm indicates higher degree of capillary condensation of N\textsubscript{2} in the mesopores compared to that of the sample C_{2x}-LDH rendering maximum ratio of mesopore to micropore surface area (S_{mesopore}/S_{micropore}) value. As micropore surface area values of C_{2x}-LDH and C_{3x/2}-LDH are comparable, the higher mesopore surface area of C_{3x/2}-LDH contributed higher BET surface area than that of the sample C_{2x}-LDH. It is worth noting that an optimum concentration of carbon with respect to LDH showed maximum S_{mesopore}/S_{micropore} rendering maximum removal efficiency.
Fig. 2 N\textsubscript{2} adsorption-desorption isotherms of C-LDH: (a) C\textsubscript{4x}-LDH, (b) C\textsubscript{2x}-LDH, (c) C\textsubscript{3x/2}-LDH and (d) C\textsubscript{x}-LDH; insets represent BJH pore size distributions (desorption) of the corresponding samples.

Table 1: Textural properties of C-LDH composite

| Sample ID | Surface area(m\textsuperscript{2}g\textsuperscript{-1}) | V\textsubscript{p}(cm\textsuperscript{3}g\textsuperscript{-1}) | Pore diameter (nm) |
|-----------|---------------------|-----------------|------------------|
|           | S\textsubscript{BET} | S\textsubscript{micropore} | S\textsubscript{mesopore} | S\textsubscript{mesopore}/S\textsubscript{micropore} |
| C\textsubscript{4x}-LDH | 758 | 611 | 147 | 0.240 | 0.857 | 1.2/3.6 |
| C\textsubscript{2x}-LDH | 520 | 424 | 96 | 0.226 | 0.628 | 3.7 |
| C\textsubscript{3x/2}-LDH | 558 | 430 | 128 | 0.297 | 0.519 | 3.9 |
| C\textsubscript{x}-LDH | 477 | 397 | 80 | 0.201 | 0.402 | 3.7 |

Note: S\textsubscript{BET} = BET surface area, S\textsubscript{micropore} = Micropore surface area, S\textsubscript{mesopore} = Mesopore surface area and V\textsubscript{p} = Total pore volume.
XPS study was performed to investigate the surface elemental states of C-LDH composites. Fig. 3a shows the presence of Al, Mg, Ca, O and C in all four samples. Fig. 3b-e reveals that the binding energies of Al2p, Mg2p, Ca2p and C1s are found to be 75.07, 50.94, 352.85 and 284.49 eV, respectively. Interestingly, the binding energies of the above elemental states shifted slightly from their standard values. It could be due to compositional variations and the change in chemical environment. In this case the sharing of elements with neighboring atoms is different in the composite samples. Binding energy of the deconvoluted peak of Al2p for four different samples represent that aluminum is present in the form of Al(OH)₃ (BE: 74.8 ± 0.1 eV) and AlOOH (BE: 75.2 ± 1.2 eV). The AlOOH is formed due to addition of excess ammonium hydroxide (NH₄OH) during the synthesis process. Interestingly, C₄ₓ-LDH sample contains mostly AlOOH instead of Al(OH)₃. On the other hand, the binding energy of Mg2p and Ca2p for four different C-LDH depicts that magnesium and calcium are present in the form of Mg²⁺ (Mg(OH)₂, BE: 50.49-51.4 eV) and Ca²⁺ (Ca(OH)₂, BE: 352.6-354.12 eV). Quantitative estimation from XPS study for all four samples is shown in Table S1, ESI†. It was observed that atomic % of carbon increases in the order of C₄ₓ-LDH > C₂ₓ-LDH > C₃ₓ/2-LDH > Cₓ-LDH. However, a little deviation in Al : Mg : Ca atomic ratio is noticed for these samples. During measurement, removal of surface oxides and contaminants by Ar⁺ ion etch could cause a number of side effects altering the composition of the upper surface.²⁹ During the study of XPS, monoatomic ion guns (Ar⁺ ions) are used to clean the surface of the sample. Sputtering with single ions may cause sample modification and several damages like ion implantation or atomic mixing, which leads to altering the composition of the investigated samples.³⁰-³² However, ICP-AES technique was applied to confirm the atomic% of Al, Mg and Ca while C% was analysed by carbon analyser. Table S2, ESI† shows C, Al, Mg and Ca contents in the sample. Based on the ICP-AES analysis, it is to be noticed that Al:Mg:Ca ratio in the sample was found to be
1:1.31-1.38:0.66-0.79 which is comparable to the experimentally calculated value of 
1:1.33:0.66.

Fig. 3 XPS spectra of C-LDH (a) full spectrum and (b) Al2p, (c) Mg2p (d) Ca2p and (e) C1s.

Fig. 4 represents the FESEM images of the samples (a) C_{4x}-LDH, (b) C_{2x}-LDH, (c) 
C_{3.5x/2}-LDH and (d) C_{x}-LDH, the high magnified images of the corresponding samples are 
shown by arrow. The LDH particles are highly agglomerated and adhered onto the surface of 
nanospherical/spheroid shaped carbon particles. In the FESEM images, nanoflake shaped 
LDH is found to be increased with increase in the concentration of LDH compared to carbon 
content of the C-LDH composites and follow in the order of C_{4x}-LDH < C_{2x}-LDH < C_{3.5x/2}- 
LDH < C_{x}-LDH. The FESEM image of porous carbon sample is shown in Fig. S2, ESI† to get 
more intuintive comparison between as-prepared porous carbon sample and carbon-LDH 
composites (Fig. 4). TEM images (Fig. 5) reveal that the particle size of carbon is in the range 
of 50-150 nm where overlapping of LDH sheets (diameter = 10-15 nm and length = 50-100 
nm) are adhered onto the surface of the carbon spheres.
Fig. 4 FESEM images of C-LDH composites: (a) C$_{4x}$-LDH, (b) C$_{2x}$-LDH, (c) C$_{3x/2}$-LDH and (d) C$_{x}$-LDH

Fig. 5 TEM images of C-LDH composites: (a) C$_{4x}$-LDH, (b) C$_{2x}$-LDH, (c) C$_{3x/2}$-LDH and (d) C$_{x}$-LDH
Adsorption study of C-LDH composite for the removal of As(V), F\(^-\), Fe(II)/Fe(III), MB and MO

Effect of contact time

Fig. 6 depicts the effect of contact time for adsorption of different pollutants: (a) As(V), (b) F\(^-\), (c) Fe(II)/Fe(III), (d) MB and (e) MO by C-LDH nanocomposites with their initial concentrations of 1.87, 3, 10, 8 and 32.7 ppm, respectively, adsorbent dosage of 0.5 g/L each at pH ~ 7 and temperature 30 °C. It shows that within 5 min, the rate of adsorption is very fast for all the contaminants. It is due to the presence of large number of vacant adsorption sites at the initial stage of adsorption. The percentage removal of the contaminants like H\(_2\)AsO\(_4\)^-\, F\(^-\), Fe(II)/Fe(III) increased steadily up to 60 min followed by their slow adsorption until the equilibrium is reached at 180 min. For the removal of MB and MO, the adsorption equilibrium is reached at 60 min. Table S3, ESI† summarizes the percentage removal of different contaminants at 5 min, 60 min and 180 min by different C-LDH composites. It is worth noting that for the removal of As(V) (Fig. 6a) and F\(^-\) (Fig. 6b), the removal capacity (%) of the sample follows in the order of C\(_{3x/2}\)-LDH > C\(_x\)-LDH > C\(_{2x}\)-LDH > C\(_{4x}\)-LDH. The maximum removal % of As (V) and F\(^-\) for the sample C\(_{3x/2}\)-LDH could be attributed to the high mesopore : micropore surface area ratio (Section 3.1) and also containing significant amount of LDH content compared to carbon content. However, the sample C\(_x\)-LDH having maximum amount of LDH shows less removal % compared to that of C\(_{3x/2}\)-LDH, corroborating to less mesopore : micropore surface area. On the other hand, for the removal of Fe(II)/Fe(III), MB and MO, the trend of adsorption % for the samples is in the order of C\(_{4x}\)-LDH > C\(_{2x}\)-LDH > C\(_{3x/2}\)-LDH > C\(_x\)-LDH which is in tune with the same order of carbon content and total surface area. In this case both the carbon content and surface area play the significant role for adsorption efficiency. Interestingly, for the adsorption of anionic
(inorganic) water contaminants such as As (V) (in the form of H$_2$AsO$_4^-$ and HAsO$_4^{2-}$) and Fe(II)/Fe(III) LDH plays a favorable role for their adsorption. However, for the adsorption of cationic inorganic water contaminant like Fe(II)/Fe(III) and organic dye (MB and MO), highly porous carbon material is the best choice.

It is inferred that for C-LDH based nanocomposite as adsorbent, the nature of the adsorbents and their relative concentration in the composite, total surface area and mesopore : micropore surface area ratio synergistically effect for the removal of different inorganic and organic based water contaminants.

![Graphs showing removal vs time for different C-LDH composites](image)

**Fig. 6** Percentage (%) removal vs time plot of different C-LDH composites (C$_{4x}$-LDH, C$_{2x}$-LDH, C$_{3x/2}$-LDH and C$_x$-LDH) for the adsorption of (a) As(V), (b) F$^-$, (c) Fe(II)/Fe(III), (d) MB and (e) MO

To determine the leading mechanism for adsorption of different pollutants, the adsorption data were fitted with different kinetic models. Fig. S3-S7 in the ESI† shows (a) pseudo-first order and (b) pseudo-second order kinetic models for the adsorption of As(V), F$^-$, Fe(II)/Fe(III), MB and MO, respectively. Table S4, ESI† summarizes the kinetics data obtained from respective curves of kinetic models. By examining the maximum R$^2$ values ($\geq$ 0.99) from these data, it is confirmed that pseudo-second order kinetic models were best
fitted. The highest adsorption capacity values were obtained from the fitted curve of the pseudo-second order kinetics. These results are found to be same with the experimental data obtained from the plot of percentage removal vs time (Fig. 6).

**Effect of adsorbate (pollutant) concentration**

The effect of initial concentrations of As(V) (2-10 ppm), F\(^-\) (5-30 ppm), Fe(II)/Fe(III) (2-20 ppm), MB (3-25 ppm) and MO (3-163 ppm) on the adsorption capacity of different C-LDH composites were studied with contact time: 180 min, adsorbent (sample) dosage: 0.5 g/L, temperature: 30 °C and pH: 6.8±0.2. Fig. 7a-e shows the change of adsorption capacity \(q_e\) with initial concentrations of different contaminants (a) As(V), (b) F\(^-\), (c) Fe(II)/Fe(III), (d) MB and (e) MO. With increasing the initial concentrations of all the pollutants, the individual adsorption capacity increased. For removal of As(V) (Fig. 7a) and F\(^-\) (Fig. 7b), the maximum adsorption capacity of C-LDH composites follows in the order of \(C_{3x/2-}\)LDH > \(C_{2x-}\)LDH > \(C_{x-}\)LDH > \(C_{4x-}\)LDH. This trend could be correlated with the carbon content in the composite, BET surface area \(S_{BET}\) and mesopore/micropore surface area ratio. With decrease in carbon content and increase in LDH concentration in the composite, the adsorption efficiency increased due to increase in anion exchange capacity of LDH. Accordingly, \(C_{4x-}\)LDH showed lowest adsorption efficacy than other composites. However, the above explanation is not hold good for the samples \(C_{3x/2-}\)LDH and \(C_{x-}\)LDH. Instead of having higher LDH content in the sample \(C_{x-}\)LDH compared to \(C_{3x/2-}\)LDH, the former showed less adsorption capacity than the latter because of lower \(S_{BET}\) and mesopore : micropore surface area ratio. It is worth mentioning that \(C_{3x/2-}\)LDH showed maximum As(V) and F\(^-\) removal capability among all the composites. It is inferred that combined effects of LDH content, BET surface area, mesopore : micropore surface area in the composites play the critical role for the removal of As(V) and F\(^-\) from water. In case of Fe(II)/Fe(III) (Fig. 7c), MB (Fig. 7d) and MO (Fig. 7e) removal, the adsorption capacity follows in the order of \(C_{4x-}\)LDH > \(C_{2x-}\)LDH > \(C_{3x/2-}\)LDH > \(C_{x-}\)LDH.
Among all the samples, C$_{4x}$-LDH showed maximum adsorption capacities due to maximum carbon content in the composite and highest BET surface area.

Fig. 7 Plot of equilibrium adsorption capacity with equilibrium concentration of different C-LDH composites (C$_{4x}$-LDH, C$_{2x}$-LDH, C$_{3x/2}$-LDH and C$_x$-LDH) for the adsorption of (a) As(V), (b) F$^-$, (c) Fe(II)/Fe(III), (d) MB and (e) MO.

In order to investigate the relationship between the adsorbate (pollutant) and adsorbent (C-LDH nanocomposite), the adsorption data is fitted with different isotherm models. Fig. S8-S12 in the ESI$^+$ represents the plots obtained from (a) Langmuir and (b) Freundlich isotherm models for the adsorption of As(V) (Fig. S8), F$^-$ (Fig. S9), Fe(II)/Fe(III) (Fig. S10), MO (Fig. S11) and MB (Fig. S12). The detailed fitting parameters of Langmuir and Freundlich models for the adsorption of pollutants are shown in Table S5, ESI$^+$. The Langmuir isotherm is expressed by the dimensionless constant separation factor R$_L$.

\[ R_L = \frac{1}{1 + K_L C_0} \]  

(3)
where $C_0$ (mg/L) is the initial concentration of pollutant and $K_L$ (L/mg) is the Langmuir constant. $R_L$ value was found in the range of 0 to 1 which indicates the favorable adsorption at room temperature. The linear form of Langmuir and Freundlich isotherms are expressed as follows:

\[
\begin{align*}
\text{Langmuir isotherm:} & \quad \frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_L q_m} \\
\text{Freundlich isotherm:} & \quad \log q_e = \log K_F + \frac{1}{n_F} \log C_e
\end{align*}
\]

Where, $C_e$ (mg/L), $q_m$ (mg/g) and $q_e$ (mg/g) are the equilibrium adsorbate concentration, maximum adsorption capacity and equilibrium adsorption capacity, respectively, while $n_F$ and $K_F$ are the adsorption intensity and Freundlich constant, respectively. The $n$ values obtained from Freundlich isotherms are found greater than 1 signifying high affinity between adsorbate and adsorbent indicating chemisorption process.\(^{33}\) Langmuir isotherm model is fitted better for the adsorption of As(V), F\(^-\), Fe(II)/Fe(III) and MB as confirmed by the $R^2$ values while Freundlich model is fitted well for the adsorption of MO. The maximum adsorption capacity values for As(V) and F\(^-\) removal are found to be 22.37 and 20.40 mg/g, respectively using C\(_{3x/2}\)-LDH, while the sample C\(_{4x}\)-LDH showed highest removal efficiencies of 80.00, 122.10 and 328.95 mg/g for the adsorption of Fe(II)/Fe(III), MB and MO, respectively at room temperature.

**Effect of adsorbent dose**

The effect of sample dose on the adsorption capacity of different C-LDH composites for the removal of different water contaminants (Initial concentration of As(V): 10.76 ppm, F\(^-\): 25.2 ppm, Fe(II)/Fe(III) : 18.48 ppm, MB : 25 ppm and MO: 25 ppm) were studied with contact time : 180 min, temperature: 30 °C and pH: 6.8±0.2. Fig. 8a-e shows the change of removal % of (a) As (V), (b) F\(^-\), (c) Fe(II)/Fe(III), (d) MB and (e) MO by changing the different adsorbent doses. For the adsorption of As(V), removal efficiency increased sharply up to the adsorbent dose of 0.5 g/L followed by slow increase up to adsorbent dose of 1.2 g/L (Fig. 8a).
The adsorbent dosage 0.5 g/L is found to be the optimum concentration for higher removal efficiency (93% for C_{3x/2}-LDH) and maximum adsorption capacity (20.37 mg/g for C_{3x/2}-LDH). Higher dose of adsorbent signifies higher surface area providing larger number of active binding sites for the contaminant ions. Consequently, adsorption efficiency increased with increasing adsorbent amount, and stabilization of adsorption is obtained after a certain concentration of adsorbent which could be due to overlapping of active sites at higher dose.

For fluoride removal, with increase in adsorbent dose from 0.2 g/L to 1.3 g/L, the rate of removal of fluoride ion is increased. For the samples, C_{4x}-LDH, C_{2x}-LDH, C_{3x/2}-LDH and C_{x}-LDH, the % removal increases from 10 to 24 %, 15.5 to 36.5 %, 26.5 to 53 %, 22.6 to 48.3 %, respectively with the increase in adsorbent dose. It is found that the maximum adsorption capacity value (q_m) reduced with increase in adsorbent concentration. As an example, the sample C_{3x/2}-LDH with dose 0.2 g/L showed q_m and % removal as 25.45 mg/g and 20.2 %, respectively while with increase in adsorbent dose as 1.3 g/L, the respective values were found to be 10.27 mg/g and 53 %. To get better adsorption capacity and removal efficiency, the optimum adsorbent dose of 0.5 g/L was used for further study for fluoride removal.

For removal of Fe(II)/Fe(III), the above phenomenon was observed i.e., with increasing the adsorbent dose, % removal of Fe(II)/Fe(III) increases significantly. For the sample C_{4x}-LDH with the adsorbent dose of 0.5 g/L, the % removal was found to be 95.6 % while it was ~88 % for other three samples. Adsorption capacity enhances with increase in doses of the adsorbent. To achieve highest adsorption capacity (q_m) with good removal efficiency, optimum adsorbent dose of 0.5 g/L was used for further study for Fe(II)/Fe(III) removal.

Similarly, for adsorption of MB (cationic dye) and MO (anionic dye), adsorption efficiency is increased with increasing adsorbent dose up to 1.0 g/L. Optimum removal
efficiency with better adsorption capacity value is obtained for the adsorbent dose of 0.5 g/L.

This dose amount of 0.5 g/L is used for further study for removal of MB and MO.

![Graphs showing adsorption efficiency](image)

**Fig. 8** Effect of adsorbent dose for adsorption of (a) As(V), (b) F⁻, (c) Fe(II)/Fe(III), (d) MB and (e) MO on C-LDH composites.

**Effect of pH**

The solution pH has significant effect for uptake of pollutants as it determines the surface charge of the adsorbent, chemical nature of the adsorbate and degree of ionization. The effect of pH in the range of 3-9 on removal of pollutants (As(V): 10.76 ppm, F⁻: 25.2 ppm, Fe(II)/Fe(III): 18.48 ppm, MB: 25 ppm and MO: 25 ppm) was studied by using optimum adsorbent dose (0.5 g/L) of composites equilibrated for time of 180 min, temperature 30 °C and pH 6.8±0.2. To study the pH effect on adsorption of different pollutants, the concentrations of As (V), F⁻, Fe(II)/Fe(III), MB and MO were taken as 10.76, 25.2, 18.48, 25 and 25 mg/L, respectively. **Fig. 9a-e** shows the pH effect for removal of different pollutants (a) As(V), (b) F⁻, (c) Fe(II)/Fe(III), (d) MB and (e) MO. Before studying the effect of pH on removal efficiency, the point of zero charge for all samples was estimated by mass titration process developed by Noh and Schwarz. The values of pH(PZC) (point of
zero charge) were found to be 7.4, 7.6, 7.78 and 8.01 for the sample C_{4x}-LDH, C_{2x}-LDH, C_{3x/2}-LDH and C_{x}-LDH, respectively. At lower pH, the surface of the samples is positively charged and showed higher adsorption due to electrostatic interaction between positively charged C-LDH composite and the negatively charged pollutants like H_2AsO_4^-, F^-, MO. However, for the removal of Fe(II)/Fe(III) and cationic dye (MB), higher adsorption occurs at pH ≥ 7. It is worth mentioning that at pH ≥ 7, the surface negative charges on the C-LDH composites increased which in turn enhance the adsorption capacity of Fe(II)/Fe(III) and MB via electrostatic attraction.

The rate of adsorption for removal of As(V) was found to be higher at lower pH followed by a decreasing trend with increase in pH of the solution (Fig. 9a). The decrease of adsorption percentage at higher pH is due to electrostatic repulsion between negatively charged H_2AsO_4^- in solution and OH^- ions. Reversely, at lower pH strong columbic interaction between negatively charged H_2AsO_4^- and positive charged C-LDH surface plays a key role for adsorption of arsenic. With increasing pH of the solution, % adsorption decreased by 25.9, 30.8, 7.8 and 32.45 % for the sample C_{4x}-LDH, C_{2x}-LDH, C_{3x/2}-LDH and C_{x}-LDH, respectively.

The sorption efficiency for removal of F^- was found to be higher at lower pH range than basic pH range (Fig. 9b). The reduced adsorption efficiency at basic pH is due to the electrostatic repulsion between negatively charged C-LDH surfaces and F^- ions. The highest F^- removal was observed at around pH 5 for all the C-LDH samples. Because of high anion exchange capacity and high surface-active binding sites, C_{3x/2}-LDH showed high removal efficiency.

The pH effect on the adsorption of Fe(II)/Fe(III) is seemed to be increased with increase the pH of the solution (Fig. 9c). The maximum adsorption was observed in the pH
range of 4.5 to 7. The pH < 4.5, the adsorption capacity is decreased due to competition between H\(^+\) ions and positively Fe(II)/Fe(III) ions. When the pH of the surrounding medium increased, negative charges on the surface of C-LDH composite increased, leading to greater electrostatic interaction between adsorbent and Fe(II)/Fe(III) ions. However, further increase of pH in the solution (pH > 7) led to the precipitation via formation of hydroxide complexes such as Fe(OH)\(_2\), Fe(OH)\(_3\) etc. Therefore, all adsorption experiments for removal of iron is conducted at pH less than 7 to avoid the effect of precipitation confirming Fe(II)/Fe(III) ions removal by adsorption only.

To study the effect of pH on the adsorption of MB, the electrostatic repulsion between cationic dye MB and the positively charged C-LDH surface inhibits the rate of adsorption of MB at lower pH. However, the adsorption efficiency was found to be significantly increased at pH 7 (Fig. 9d). The reason behind that the amino group of MB becomes free from protonation at neutral pH leading to electrostatic attraction with vacant adsorption sites of C-LDH composites.\(^{35}\) Further, the adsorption of MB decreases at alkaline pH. The negative charge on the C-LDH surface increases at higher pH causing repulsion between oxyanions of MB dye (formed through the complex formation) and the adsorbent.\(^{36}\)

The adsorption capacity of methyl orange on the surface of C-LDH was found to be higher at lower pH (Fig. 9e) which is due to electrostatic attraction between the anionic dye MO and positively charged C-LDH composites. With increase the pH of the solution (>7), the adsorption capacity reduces due to electrostatic repulsion between anionic dye, MO and negatively charged surface of composites.
The effect of pH on adsorption capacity of different C-LDH composites (C_{4x}-LDH, C_{2x}-LDH, C_{3x/2}-LDH and C_{x}-LDH) for the adsorption of (a) As(V), (b) F^−, (c) Fe(II)/Fe(III), (d) MB and (e) MO.

**Effect of temperature**

To understand the temperature effect on sorption capacity of C-LDH for As(V), F^−, Fe(II)/Fe(III), MB and MO removal, different temperatures were selected in the range of 30 °C (303 K) to 60 °C (333 K) with initial As(V), F^−, Fe(II)/Fe(III), MB and MO concentrations of 10.76, 25.2, 18.48, 25 and 25 mg/L, respectively with contact time: 180 min, and pH: 6.8±0.2. Fig. 10 shows the effect of % removal efficiency of C-LDH nanocomposites for removal of different contaminants on C-LDH composites with temperature. Rate of reaction between adsorbent and adsorbate during adsorption process is influenced by the temperature. With increase in temperature, diffusion of adsorbate molecules increases through the external laminar layer into the pores of the adsorbent resulting change of equilibrium adsorption capacity. For both cases, adsorption capacity is increased by increase in temperature up to 40 ± 2 °C (313 ± 2 K). Further increase in temperature, the rate of removal efficiency remains almost constant for all C-LDH
composites. This could be due to disruption of active binding sites at adsorbent or destruction of adsorbate molecules or increasing desorption tendency of contaminant ions from interface of the solution.  

\[ C_{3x/2}^- \text{-LDH} \] showed maximum removal efficiency as compared to other C-LDH samples. For \[ C_{3x/2}^- \text{-LDH} \], adsorption capacity increases from 22.30 mg/g to 23.14 mg/g (for As(V)), 19.15 mg/g to 19.99 mg/g (for \( F^- \)), 35.25 to 37.36 mg/g (for Fe(II)/Fe(III)), 41.79 to 43.95 mg/g (for MB) and 48.46 to 48.83 mg/g (for MO) with increase in temperature from 30 ± 2 °C (303 ± 2 K) to 40 ± 2 °C (313 ± 2 K).

**Simultaneous removal of As(V), \( F^- \), Fe(II)/Fe(III), MB and MO**

Due to higher removal efficiency of the sample \( C_{3x/2}^- \text{-LDH} \) for the adsorption of water contaminants, particularly, As(V) and \( F^- \), it was used for simultaneous removal of As(V), \( F^- \), Fe(II)/Fe(III), MB and MO from water. In this study, the experiments were carried out by changing concentrations of adsorbate (Fig. 11 a) (keeping sample dose 3.75 g/L) and adsorbent dosage (Fig. 11 b) (keeping pollutant concentration of 5 mg/L) with contact time.
180 min, at pH 6.8±0.2, temperature 30 °C each. The percentage removal of different pollutants simultaneously by using C_{3x/2}-LDH composite was compared with the as-prepared carbon sample and LDH sample (Fig. S13 in the ESI†). It is worth noting that the similar experimental procedure was used to synthesize the LDH sample in absence of carbon. By changing the pollutant (adsorbate) concentrations, it was noticed that % removal for fluoride decreased from 86.5 % to 75 % with increase in adsorbate concentrations from 5 to 20 mg/L. However, no significant changes in % removal of As(V) and dye (MB+MO) with increase in adsorbate concentration, it was ~99% and > 99% respectively for the adsorbate concentration in the range of 5-20 mg/L. For the adsorption of Fe(II)/Fe(III), the % removal increased from 95 % to 99 % by changing the adsorbate concentration from 5 to 10 mg/L followed by no significant change up to the adsorbate concentration of 20 mg/L. Presence of co-existing ions of As(V) in the solution reduced the fluoride adsorption. It was reported that after adsorption of arsenate (As(V)), the zeta potential of the double layer on surface is decreased significantly to negative values, as a result fluoride adsorption on this negatively charged surface is inhibited.\textsuperscript{15, 39} However, removal of As(V) was not affected significantly in the presence of fluoride on the surface of the C-LDH composite. Generally, fluoride ions are adsorbed by ligand exchange mechanism \textit{via} exchange of surface OH\textsuperscript{-} with F\textsuperscript{-}. Therefore, fluoride adsorption should not change the charge and the potential of the surface of C-LDH during As(V) adsorption. Fe(II)/Fe(III) is responsible for enhanced adsorption capacity of As(V) \textit{via} formation of arsenate-Fe(II)/Fe(III) complexes.\textsuperscript{40} In presence of excess iron, this complex formation tendency is increased and arsenate is precipitated with excess iron. Consequently, removal of iron simultaneously facilitates the arsenic removal from water.
Fig. 11 (a) Percentage removal of F⁻, As(V), Fe(II)/Fe(III), mixed dye (MB and MO) using composite C₃ₓ/₂-LDH by changing the pollutant concentration, (b) Percentage removal of F⁻, As(V), Fe(II)/Fe(III), mixed dye (MB and MO) using composite C₃ₓ/₂-LDH by changing the sample doses.

Fig. 11(b) shows that with increase in adsorbent (sample) dose from 0.625 to 3.75 g/L, the adsorption efficacy (%) slightly increased from ~98 % to ~99.9 % for the removal of As(V) and dye (MB+MO). However, there is a steady increase in % removal of fluoride and Fe(II)/Fe(III) from 37 % to 86 % and 85 % to 95 %, respectively with increase in sample dose from 0.625 to 3.75 g/L. Increase in sample dose increases the number of active adsorption sites which, in turn, enhances the adsorption capacity towards the removal of pollutants.

**Effect of coexisting ions**

Fig. 12 shows the effect of different co-existing ions like Cl⁻, NO₃⁻, SO₄²⁻, PO₄³⁻ on removal of contaminants (As(V), F⁻, Fe(II)/Fe(III), MB and MO). The initial concentration of each ion was kept at 5 mg/L at pH 7 with contact time of 180 min at 30 °C. It was found that the removal efficiency remains almost same for the adsorption of As(V), Fe(II)/Fe(III), MB and MO in presence of competing ions. However, the percentage removal of F⁻ was decreased in the presence of competing ions. The presence of Cl⁻, NO₃⁻, SO₄²⁻, PO₄³⁻ enhances the coulombic repulsion forces and compete with F⁻ ions for the active adsorption sites of C-
LDH nanocomposites. As a result, removal efficiency for the adsorption of \( \text{F}^- \) reduced in the presence of coexisting ions in water sources.

![Graph showing % Removal for different substrates](image)

**Fig. 12** Effect of coexisting ions on adsorption of As(V), \( \text{F}^- \), Fe(II)/Fe(III), MB and MO by \( C_{3x/2}-\text{LDH} \) nanocomposites.

**Study of regeneration and recyclability**

The C-LDH composite sample was regenerated by washing with Millipore water and mild NaOH solution (0.1 N) for multiple times to remove the adsorbed pollutants. Recyclability test was performed using the regenerated samples up to four consecutive cycles. It was found that percentage removal remained almost same up to 4\(^{\text{th}}\) cycles for adsorption of iron, MB and MO. However, the removal efficiency was found to be decreased from 2\(^{\text{nd}}\) cycle for the adsorption of arsenate and fluoride (Fig. 13).
Fig. 13 Percentage removal for the adsorption of As(V), F\(^-\), Fe(II)/Fe(III), MB and MO on C\(_{3x/2}\)-LDH in four consecutive cycles.

**Mechanism of adsorption**

The synthesized composite material C-LDH showed significant adsorption capacity for removal of both inorganic (metal cations, anions and oxyanions) and organic (dyes) water pollutants due to synergistic effect of carbon and LDH content. The porous carbon in LDH structure helps improve surface area, chemical stability and oxygen containing surface functional groups. As a consequence, effective numbers of active adsorption sites are increased rendering enhanced adsorption. Adsorption is occurred via electrostatic interaction, ion exchange and complex formation mechanisms. Electrostatic attraction of arsenate ions with C-LDH nanocomposite and complex formation with iron are the major removal mechanism for arsenic. Fig. S14 in the ESI\(^+\) shows the FTIR study (before and after adsorption of water pollutants) to understand the adsorption mechanism. The appearance of peak at around 794 cm\(^{-1}\) signifies As-O bond formation after adsorption of As(V) onto C\(_{3x/2}\)-LDH. The reduction of the peak intensity at around 1379 cm\(^{-1}\) of the intercalated anions (NO\(_3^-\)) signifies the partial replacement of interlayer anions of LDH with arsenate and fluoride. On the other hand, it is observed that after adsorption of contaminants onto LDH structure, the intensity of absorption band at around 654 cm\(^{-1}\) due to Mg-O bond is found to be diminished. It demonstrates that Mg\(^{2+}\) ions are partially exchanged by the pollutant ions.
during adsorption. Methyl orange is adsorbed on C-LDH composites by electrostatic attraction between the dye molecules (MB+MO) and LDH. Additionally, the carbon in LDH facilitates the formation of H-bonding between anionic dye MO and carbon. Therefore, the cooperative contribution of electrostatic attraction and hydrogen bonding between dye molecules and C-LDH composites is advantageous for the improvement of adsorption capacity. Fig. 14 represents the tentative adsorption mechanism for removal of pollutants using C-LDH composites. Four different C-LDH composite samples were synthesized by varying the concentration of carbon with respect to LDH. From the removal study, it was found that the samples C₄ₓ-LDH (C = 0.06 mol) showed higher adsorption of organic dyes and Fe(II)/Fe(III) compared to As(V) and F⁻ removal. However, with decrease in the mols of carbon with respect to LDH, reduced the adsorption of organic dyes and increased the adsorption of As(V) and F⁻. For example, C₁ₓ-LDH showed higher As(V) and F⁻ removal efficiency. From this findings, it can be mentioned that the porous structure of carbon is responsible for the removal of organic dyes and Fe(II)/Fe(III), whereas the interlayer space of the LDH structure adhered outside of the carbon sphere is utilized for removal of As(V) and F⁻. The simultaneous removal of pollutants can also be explained by the co-adsorption mechanism. The negatively charged arsenate and fluoride could improve the adsorption of positively charged dye, MB through electrostatic interaction. On the other hand, the positively charged Fe(II)/Fe(III) facilitates the adsorption of negatively charged MO by electrostatic interaction. At the same time, N-containing groups of the adsorbed MB and MO could enhance the adsorption sites for arsenate, fluoride and iron. Therefore, the simultaneous removal mechanism could be attributed to the (i) presence of N-containing groups on the adsorbed MB and MO and (ii) electrostatic interaction through the arsenate, fluoride and iron.
C-LDH nanocomposite was prepared for simultaneous removal of As(V), F⁻, Fe(II)/Fe(III), MB and MO from water. Textural properties C-LDH composites indicates that BET surface area ($S_{BET}$) values follows in the order of $C_{4x}$-LDH (758 m²/g) > $C_{3x/2}$-LDH (558 m²/g) > $C_{2x}$-LDH (520 m²/g) > $C_{x}$-LDH (477 m²/g), while $S_{mesopore}/S_{micropore}$ values follows in the trend of $C_{3x/2}$-LDH > $C_{4x}$-LDH > $C_{2x}$-LDH > $C_{x}$-LDH. The compositional variation of C : LDH has an important role in changing the textural behaviours. The maximum removal % of As(V) and F⁻ for the sample $C_{3x/2}$-LDH could be attributed to the high mesopore : micropore surface area ratio. For the removal of Fe(II)/Fe(III), MB and MO, the trend of adsorption % for the samples is in the order of $C_{4x}$-LDH > $C_{2x}$-LDH > $C_{3x/2}$-LDH > $C_{x}$-LDH which is in tune with the same order of carbon content and surface area. The maximum adsorption capacity values for As(V) and F⁻ removal are found to be 22.37 and 20.40 mg/g, respectively using $C_{3x/2}$-LDH, while the sample $C_{4x}$-LDH shows highest adsorption efficacy of 80.00, 122.10 and 328.95 mg/g for the adsorption of Fe(II)/Fe(III), MB and MO, respectively at room temperature. The nature of the adsorbents and their relative concentration in the composite, total surface area and mesopore : micropore surface area ratio synergistically effect for the
removal of different inorganic and organic based contaminants from water. For simultaneous removal of As(V), F\(^{-}\), Fe(II)/Fe(III) and dye (MB and MO) with concentration 5 mg/L each from water, the composite material C\(_{3x/2}\)-LDH having 3.75 g/L dosage shows percentage removal as 99.8 %, 86.5 %, 94.8 % and 99.99 %, respectively at pH 6.8±0.2 and temperature 30 °C. The present work is significant for development of C-LDH nanocomposites for the removal of toxic water pollutants like As(V), F\(^{-}\), Fe(II)/Fe(III) and different cationic and anionic dyes from waste water via an eco-friendly process.

Conflicts of interest

One patent applications covering this work have been filed by CSIR-Central Glass & Ceramic Research Institute (Indian patent application file no. 202011034408, dated 11.08.2020)

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