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Support Information

Adsorption of nitrate from water by quaternized chitosan wrinkled microspheres@MgFe-LDHs core-shell composite

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11 **1. Preparation of materials and characterization**

12 **1.1. Preparation of GCS**

13 First, 250 ml liquid paraffin was added into a 500 ml round bottom flask, and a
14 certain amount of Span80 was added. Then, 25 ml chitosan acetic acid solution (1 g
15 chitosan was dissolved in 3% (V/V) acetic acid solution, and the suspension was
16 uniformly dispersed by ultrasound) was added slowly at 500 rpm and kept in a constant
17 temperature water bath at 50°C for 3 h. After that, 0.8 ml glutaraldehyde was added
18 slowly and water bath was continued for 4 h. The liquid in the round bottom flask was
19 centrifugally washed three times with petroleum ether and isopropanol, and dried in an
20 oven at 60°C, GCS was successfully synthesized.

21 **1.2. Preparation of GCS@CTA**

22 0.4 g GCS and 10 ml isopropanol were added to the three-necked flask, then heated
23 to 50°C with stirring, swelled for 1 h, slowly added dropwise with 2 mL of CTA, and
24 then reacted at 70°C for 7 h. Finally, the liquid in the three-necked flask was centrifuged
25 and washed three times with methanol and ethanol and dried in an oven to obtain the
26 GCS@CTA.

27 **1.3. Preparation of MgFe-Cl-LDHs**

28 $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ were added into 50 mL deionized water, and the
29 mixed metal salt solution of 0.06 mol L^{-1} was prepared with the molar ratio of $[\text{Mg}^{2+}]/$
30 $[\text{Fe}^{3+}]$ of 2:1, 3:1, 4:1, respectively. Then added 1 mol L^{-1} NaOH solution under
31 vigorous stirring to ensure that the pH value of the solution was between 9-10. Then
32 the solution was transferred to a stainless steel autoclave and put in the oven at 120°C
33 for 12 h. Finally, deionized water and ethanol were used for centrifugal washing until
34 the solution was neutral, and the products were dried in an oven to obtain 2 MgFe-Cl-
35 LDHs, 3 MgFe-Cl-LDHs, and 4 MgFe-Cl-LDHs, respectively.

36 **1.4. Preparation of GCS@CTA @MgFe-LDHs**

37 Add 0.2 g of GCS@CTA to 15 mL of deionized water and stir for 30 min obtained
38 solution A. Solution B was obtained by adding 0.4879 g of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ and 0.1622 g
39 of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ to 50 mL of deionized water. Subsequently, solution B was added
40 dropwise to solution A with constant stirring, and 1 mol/L NaOH was used to ensure

41 the pH value was between 9-10. The mixed solution was transferred to a stainless steel
42 autoclave and reacted in an oven at 120 °C for 12 h. Finally, deionized water and
43 ethanol were used for centrifugal washing until the solution was neutral, and the product
44 was dried in a 60 °C oven to obtain the GCS@CTA@MgFe-LDHs.

45 **1.5. Characterization**

46 The crystallographic of samples were measured by X-ray diffraction (XRD-D8
47 Advance, Bruker, Cu K α radiation $\lambda=1.5406$ nm). FT-IR spectroscopy (Spectorn One,
48 Perkin Elmer) was selected to verify the samples. Micromorphological of samples were
49 characterized by scanning electron microscopy (Hitachi S-4800). The Brunauer-
50 Emmett-Teller (BET) surface areas of the samples were measured by N₂ adsorption on
51 AUTOSORB-1, and the plot of the pore diameter distribution was measured by Barrett-
52 Joyner-Halenda (BJH) method. The elemental composition of samples were measured
53 by X-ray photoelectron spectroscopy (XPS). The zeta potential was measured by Zeta
54 Sizer Nano-ZS system (Malvern, United Kingdom).

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56 Table S1. Comparison of maximum nitrate adsorption capacities (q_m , mg g⁻¹) by
 57 different adsorbents reported in prior literatures.

Adsorbents	Dosage	pH	q_m (mg/g)	References
Polypyrrole-modified plastic-carbon	15 g/L	2-11	45.18	[1]
ZnCl ₂ -AC	20 g/L	4	5.53	[2]
Carboxymethyl β -cyclodextrin immobilized on hydrated lanthanum	0.5 g/L	3-11	37.50	[3]
La ₂ O ₃ AM@CS composite beads	2 g/L	3-11	27.84	[4]
Zr/quaternary ammonium powder with polyvinylidene fluoride	--	--	9.66	[5]
Zr cross-linked chitosan assisted bentonite	2 g/L	3-11	23.89	[6]
modified cellulose from Corn stalks	0.1 g	6.8	13.61	[7]
Chitosan grafted quaternized resin	2 g/L	2-11	34.5	[8]
GCS@CTA@MgFe-LDHs	0.4 g/L	4-11	71.5	This study

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61 Table S2. Concentrations of Mg^{2+} and Fe^{3+} leached from GCS@CTA@MgFe-LDHs

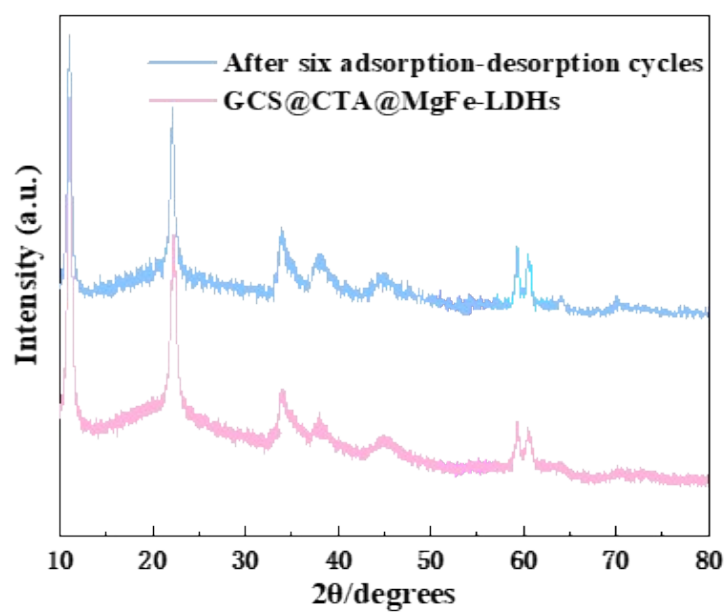
62 extracted solution at pH 4

Sample	Mg^{2+} (mg L^{-1})	Fe^{3+} (mg L^{-1})	pH ($T = 298\text{K}$)
GCS@CTA@MgFe-LDHs	0.128	0.073	4 ± 0.5

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67 Fig.S1 The XRD of GCS@CTA@MgFe-LDHs before and after six adsorption-
68 desorption cycles

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