Supporting Information for

Charge-switchable zwitterionic nanomagnets for wastewater remediation

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Determination of isoelectric point (IEP):

The isoelectric point of the NTA@SPIONs was determined by recording the zeta potential of the particle dispersions over a range of pH from 3 to 7. The medium pH was adjusted by the addition of 0.1M HCl. The obtained zeta potential was plotted against pH. The pH at which the plot intersects $\xi=0$ is taken as the isoelectric point.

Reusability tests:

After adsorption, the magnetically collected adsorbent material was dispersed in proper eluting mediums, followed by bath sonication, to desorb the loaded dye species. For MB, an HCL solution (pH=3) was used, and for CR, a NaOH solution (pH=11) was employed as the eluting medium. Following the elution, the adsorbent material was collected with a bar magnet. The whole eluting process was repeated twice. Finally, the adsorbent material was washed thoroughly with miliQ water and dried overnight. For the reusability test, the adsorption-desorption cycle was repeated five times. After each cycle, the supernatant was analyzed to estimate the removal efficiency of the adsorbent.



Figure S1: Large-area TEM image of NTA@SPIONs with a photograph of material obtained from a single batch (left inset) and indexed SAED pattern matching with spinel magnetite phase (right inset). HRTEM and corresponding FFT (inset) for a particle with (a) light, and (b) dark contrast. Indexed planes match with the spinel magnetite phase.



Figure S2: Hydrodynamic particle size measurement (DLS) of NTA@SPIONs immediately after preparation and after a gap of seven days.



Figure S3: Langmuir isotherm fitting for MB, CR, Cr^{3+} and $Cr_2O_7^{2-}$. Inset shows the dependency of removal efficiency on adsorbate concentration.



Figure S4: Pseudo-second order kinetics fitting for MB, CR, Cr³⁺ and Cr₂O₇²⁻.



Figure R5: PXRD of the NTA@SPIONs before and after Cr₂O₇²⁻ adsorption.



Figure R6: TEM images of the NTA@SPIONs (a) before and (b) after $Cr_2O_7^{2-}$ adsorption.



Figure R7: EDS elemental mapping of the NTA@SPIONs after $Cr_2O_7^{2-}$ adsorption at pH 4 (a) Fe, (b) Cr, and (c) O. (d) EDS spectrum of the NTA@SPIONs after $Cr_2O_7^{2-}$ adsorption.

Model	Langmuir		
Parameters	q_m	b	R ²
MB	44.98	0.32	0.99
CR	273.79	1.51	0.99
Cr ³⁺	13.31	1.29	0.98
Cr ₂ O ₇ -	4.10	0.49	0.98

Table S1: Langmuir isotherm fitting parameter for the pollutants.

Model	Pseudo-second order	
Parameters	k	R ²
MB	0.202	0.99
CR	0.052	0.99
Cr ³⁺	0.054	0.99
Cr ₂ O ₇ ⁻	0.046	0.99

Table S2: Pseudo-second order fitting parameter for the pollutants.

References:

- 1 S. Reja, M. Kumar and S. Vasudevan, *Nanoscale Adv.*, 2024, 3857–3864.
- 2 X. Zhang, P. Zhang, Z. Wu, L. Zhang, G. Zeng and C. Zhou, *Colloids Surfaces A Physicochem. Eng. Asp.*, 2013, **435**, 85–90.
- S. Yesmin, M. Mahiuddin, A. B. M. Nazmul Islam, K. M. R. Karim, P. Saha, M. A. R.
 Khan and H. M. Ahsan, *ACS Omega*, DOI:10.1021/acsomega.3c09557.
- 4 S. Technology, A. Wiryawan, R. Retnowati and R. Y. P. Burhan, 2018, 05, 37–46.
- 5 A. Lace, D. Ryan, M. Bowkett and J. Cleary, *Int. J. Environ. Res. Public Health*, , DOI:10.3390/ijerph16101803.
- T. Shigematsu, S. Gohda, H. Yamazaki and Y. Nishikawa, *Bull. Inst. Chem. Res., Kyoto Univ.*, 2007, 55, 429–440.

 A. Wiryawan, R. Retnowati, P. Burhan and S. Syekhfani, J. Enviromental Eng. Sustain. Technol., 2018, 5, 37.