

Article

A Novel Magnetic Nano-Adsorbent Functionalized with Green Tea Extract and Magnesium Oxide to Remove Methylene Blue from Aqueous Solutions: Synthesis, Characterization, and Adsorption Behavior

Wenchao Lin ^{1,2}, Yaoyao Huang ³, Shuang Liu ¹, Wei Ding ^{1,2,*}, Hong Li ^{1,*} and Huaili Zheng ^{1,2}

¹ Key Laboratory of the Three Gorges Reservoir Region's Eco-Environment, Ministry of Education, Chongqing University, Chongqing 400045, China; 13042332885@163.com (W.L.); 13658323766@163.com (S.L.); hlz6512@163.com (H.Z.)

² College of Environment and Ecology, Chongqing University, Chongqing 400044, China

³ National Research Base of Intelligent Manufacturing Service, Chongqing Technology and Business University, Chongqing 400067, China; huangyaoyao8845@163.com

* Correspondence: weiding@cqu.edu.cn (W.D.); lihong666@cqu.edu.cn (H.L.)

Text S1. Characterization techniques.

The surface morphology of the prepared materials were examined by SEM (TM4000Plus II, Hitachi, Tokyo, Japan) attached with an energy dispersive spectrometer (EDS). Prior to observation, the samples were crushed, dispersed in ethanol, dispersed by ultrasound, and deposited on copper mesh.

The phase structure and crystallinity was measured by XRD (PANalytical X'Pert Powder, Spectris Pte. Ltd., Almelo, the Netherlands) using Cu K α radiation (40 kV, 40 mA, $\lambda=1.5406$ Å). Scan range: 2 θ of 10° to 80°; Step size: 0.026°.

The infrared spectrum of prepared samples was recorded by FTIR (IRTracer100, Shimadzu, Kyoto, Japan) with the KBr pellet technique.

The composition of elements in the prepared materials was analyzed by XPS (ESCALAB250Xi, Thermo Fisher, Waltham, Massachusetts, USA). With a 400 μm spot size, 12 kV operating voltage, and 6 mA filament current, the full-spectrum scanning fluence energy was 150 eV in 1 eV steps, whereas the narrow-spectrum scanning fluence energy was 50 eV in 0.1 eV steps.

The magnetic properties were measured in a commercial PPMS DynaCool 9 (Quantum Design, San Diego, California, USA).

The point of zero charge of the catalyst was determined with zetasizer nano ZS90 apparatus (Malvern, London, UK).

The pore texture parameters and surface areas were analyzed using automatic multi-station specific surface and aperture analyzer (Quadrasorb 2MP, Quantachrome, Boca Raton, Florida, USA). The pore texture parameters were obtained from the corresponding N₂ adsorption–desorption isotherms. The pore size distribution was obtained through the classical Barrett–Joyner–Halenda method based on a modified Kelvin adsorbent. The specific surface areas were estimated by the Brunauer–Emmet–Teller method using the adsorption branch of the isotherms.

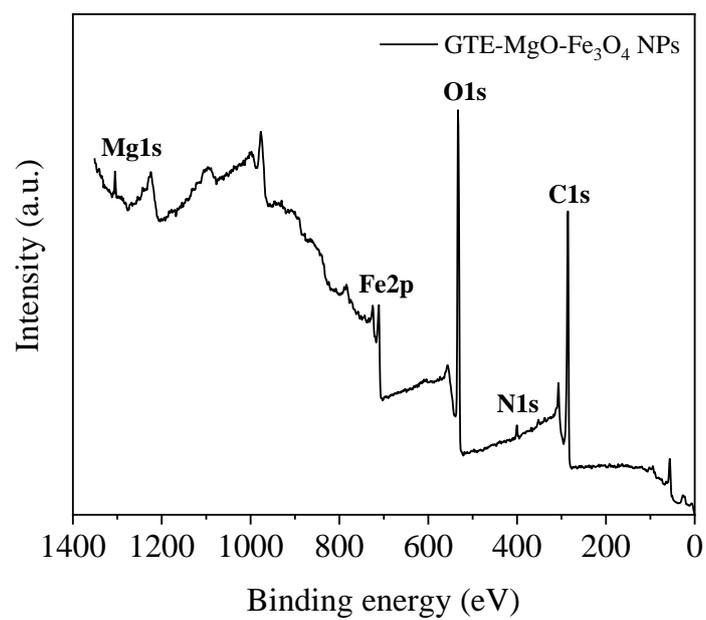


Figure S1. The XPS fully scanned spectra of GTE-MgO-Fe₃O₄ NPs.

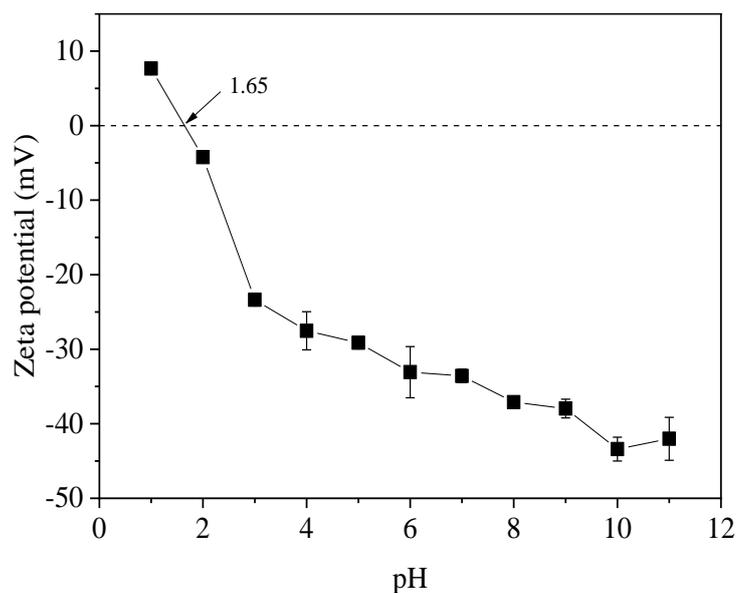


Figure S2. Zeta potential curve for GTE-MgO-Fe₃O₄ NPs in the pH range of 1-11.

Table S1. Kinetic model parameters for the MB adsorption by GTE-MgO-Fe₃O₄ NPs.

kinetic model	parameters	Initial concentration of methylene blue (mg L ⁻¹)			
		10	20	40	60
PFO	q_e (mg g ⁻¹)	32.37	62.76	107.16	126.41
	k_1 (min ⁻¹)	0.570	0.293	0.189	0.166
	R^2	0.90	0.95	0.95	0.94
PSO	q_e (mg g ⁻¹)	33.76	66.57	115.46	136.83
	k_2 (g mg ⁻¹ min ⁻¹)	0.0305	0.0068	0.0023	0.0017
	R^2	0.99	0.99	0.99	0.99

Table S2. Isotherm model parameters for the MB adsorption by GTE-MgO-Fe₃O₄ NPs.

Model	Parameters	288.15 K	298.15 K	308.15 K	318.15 K
Langmuir	q_m (mg g ⁻¹)	146.33	139.60	170.94	170.46
	k_L (L mg ⁻¹)	5.57	2.60	0.11	0.07
	R^2	0.66	0.67	0.92	0.97
Freundlich	k_F ((mg g ⁻¹)(L mg ⁻¹) ^{1/n})	95.63	86.42	33.70	26.06
	n	6.51	6.32	2.60	2.34
	R^2	0.99	0.99	0.99	0.99
Temkin	b_T (J mol ⁻¹)	157.82	154.52	83.75	80.21
	f (L mg ⁻¹)	996.05	293.97	1.96	1.05
	R^2	0.96	0.98	0.94	0.97

Table S3. Thermodynamic parameters for the MB adsorption by GTE-MgO-Fe₃O₄ NPs.

temperature (K)	pH	ΔG° (kJ mol ⁻¹)	ΔH° (kJ mol ⁻¹)	ΔS° J mol ⁻¹ k ⁻¹
288.15	6.0	-44.10		
298.15	6.0	-43.74		
308.15	6.0	-37.11	-123.62	-276.14
318.15	6.0	-37.01		

Table S4. The maximum adsorption capacities (q_{\max}) of various adsorbents for MB.

Adsorbents	q_{\max} (mg g ⁻¹)	R%	Ref
Fe ₃ O ₄ @GTPs NPs	7.25	95	[37]
γ -Fe ₂ O ₃ @GL	91.74	99	[38]
magnetic-biochar nanocomposite derived from avocado peel	62.1	-	[39]
NiFe ₂ O ₄ @GO	76.34	-	[40]
CMC-coated Fe ₃ O ₄ @SiO ₂	22.7	-	[41]
Chitosan/Fe ₃ O ₄ nanocomposite	45.37	-	[42]
Fe ₃ O ₄ @Ag/SiO ₂	128.5	99.6	[43]
TTGTW500	69.01	98.07	[44]
GTE-MgO-Fe ₃ O ₄ NPs	174.93	99.78	This work