

# **Supporting Information**

## **Hyperbranched Polyethylenimine-Tethered Multiple Emulsion-Templated Hierarchically Macroporous Poly(acrylic acid)–Al<sub>2</sub>O<sub>3</sub> Nanocomposite Beads for Water Purification**

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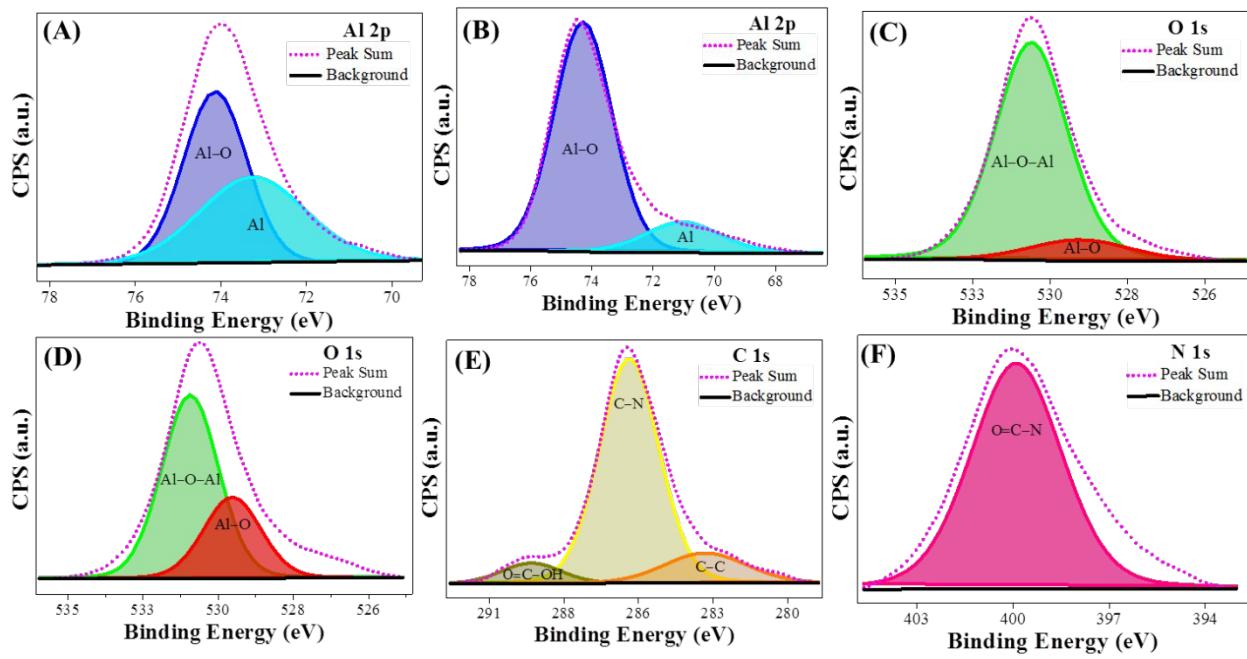
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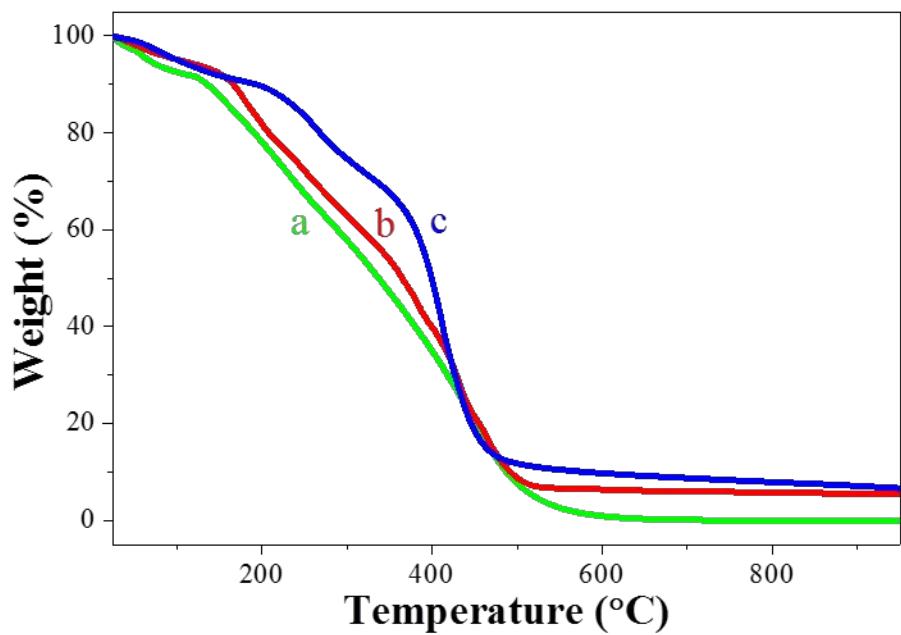
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**Table S1.** General characteristics of AO NPs and PAA and PAA–AO–PEI NC beads.

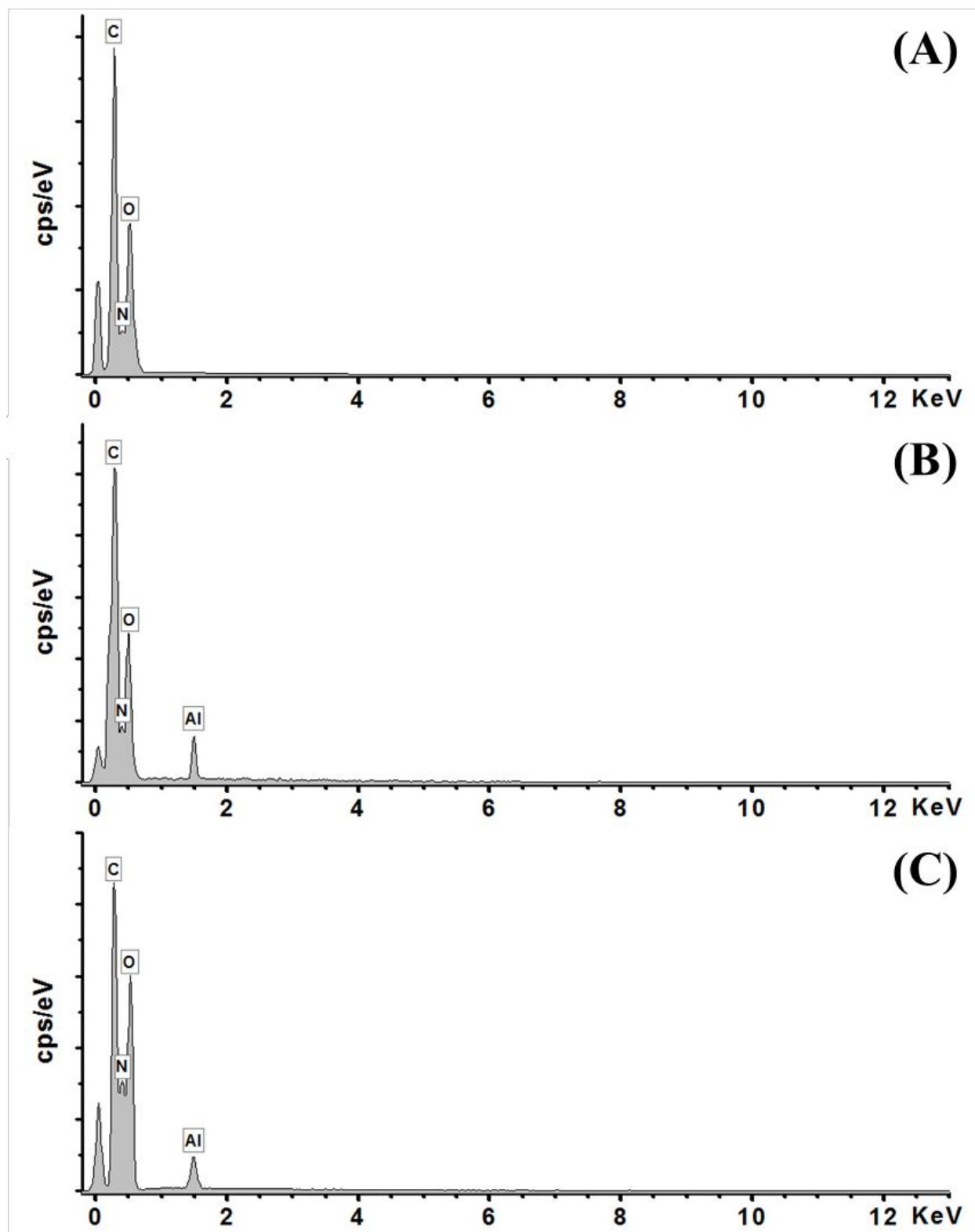
Characteristic	AO	PAA–AO	PAA–AO–PEI
size of the adsorbent	<50 nm	—	2.02 ± 0.06 mm
BET surface area (m <sup>2</sup> /g)	>40	—	5.42
zeta potential	33.30 (pH = 2) 42.20 (pH = 6)	22.60 (pH = 3) 9.50 (pH = 6)	38.30 (pH = 3) 18.80 (pH = 5)
leaching of AO (by ICP-OES)	—	not detected	not detected



**Figure S1.** High-resolution Al 2p, O 1s, C 1s, and N 1s XPS spectra of (A,C) AO NPs and (B,D,E,F) PAA–AO NC beads.



**Figure S2.** TGA curves of (a) PAA, (b) PAA–AO, and (c) PAA–AO–PEI NC beads.



**Figure S3.** EDX spectra of (A) PAA, (B) PAA–AO, and (C) PAA–AO–PEI NC beads.

**Table S2.** Parameters of Langmuir and Freundlich isotherm models for the adsorption of Cr(VI) and CR onto the AO NPs and PAA–AO and PAA–AO–PEI NC beads.

Type of Isotherm	Parameters	AO NPs		PAA–AO NC bead		PAA–AO–PEI NC bead	
		Cr(VI)	CR	Cr(VI)	CR	Cr(VI)	CR
Langmuir	$q_{\max}$ (mg/g)	95.52	19.03	128.52	25.83	141.09	37.04
	$b$ (L/mg)	$7.11 \times 10^{-3}$	$4.85 \times 10^{-2}$	$2.95 \times 10^{-3}$	$3.79 \times 10^{-2}$	$2.15 \times 10^{-3}$	$2.39 \times 10^{-2}$
	$R_L$	0.97	0.83	0.99	0.87	0.99	0.91
Freundlich	$R^2$	0.98	1.00	0.98	1.00	0.99	0.99
	$K_f$	5.28	3.35	9.06	5.39	10.38	7.97
	$1/n$	0.47	0.31	0.44	0.26	0.44	0.24
	$R^2$	0.97	0.90	0.95	0.99	0.97	0.98

**Table S3.** Parameters of pseudo-first-order and pseudo-second-order kinetics models for the adsorption of Cr(VI) and CR on AO NPs and PAA–AO and PAA–AO–PEI NC beads.

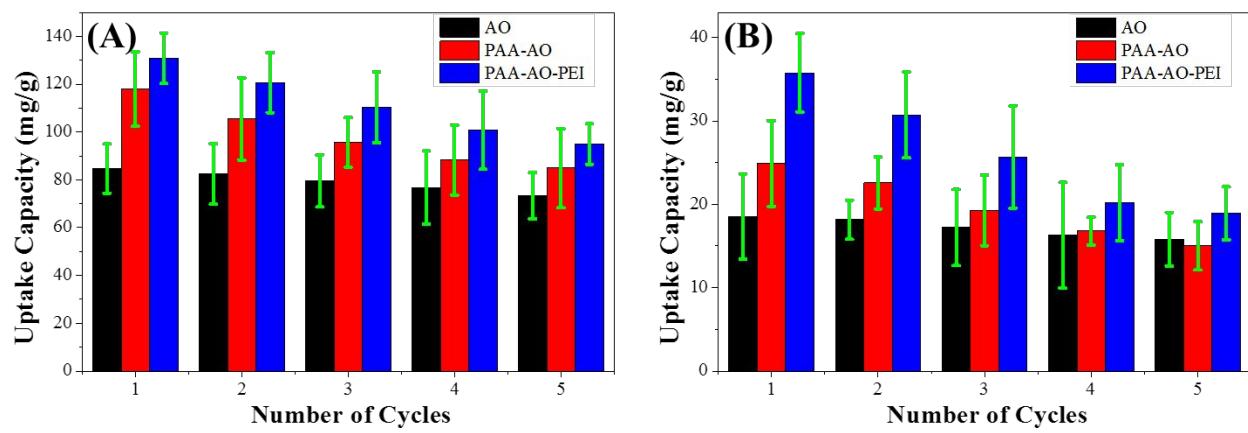
Order of Reaction	Parameters	AO NPs		PAA–AO NC bead		PAA–AO–PEI NC bead	
		Cr(VI)	CR	Cr(VI)	CR	Cr(VI)	CR
pseudo-first-order	$q_e$ (mg/g)	84.70 (E) 66.19 (T)	18.54 (E) 12.90 (T)	117.92 (E) 101.85 (T)	24.89 (E) 16.71 (T)	130.81 (E) 97.26 (T)	35.76 (E) 29.81 (T)
	$k_1$ (min <sup>-1</sup> )	$3.94 \times 10^{-02}$	$2.64 \times 10^{-02}$	$1.85 \times 10^{-02}$	$1.15 \times 10^{-02}$	$1.92 \times 10^{-02}$	$1.36 \times 10^{-02}$
	$R^2$	0.98	0.95	0.97	0.99	0.99	0.98
pseudo-second-order	$q_e$ (mg/g)	84.70 (E) 95.80 (T)	18.54 (E) 22.19 (T)	117.92 (E) 132.94 (T)	24.89 (E) 27.65 (T)	130.81 (E) 148.26 (T)	35.76 (E) 40.98 (T)
	$k_2$ (g mg <sup>-1</sup> min <sup>-1</sup> )	$3.23 \times 10^{-04}$	$9.78 \times 10^{-04}$	$2.30 \times 10^{-04}$	$1.10 \times 10^{-03}$	$2.28 \times 10^{-04}$	$5.76 \times 10^{-04}$
	$R^2$	1.00	1.00	1.00	0.99	1.00	0.99

E stands for experimental and T stands for theoretical values.

**Table S4.** The Cr(VI) adsorption capacities comparison of AO NPs and PAA–AO and PAA–AO–PEI NC beads with those of the recently reported benchmark adsorbents.

Material	Cr(VI) Adsorption Capacity (mg/g)	Operational Parameters					Ref.
		g/L of adsorbent	$C_0$ (ppm)	pH	h		
Fe <sub>3</sub> O <sub>4</sub> -CDI-IL MNPs	2.60	3	0.20–1	3	3	1	
PPy@poly(AA-co-MBA)	15.55	1.63	200	2	20/6	2	
regenerated cellulose membrane	18.13	1	20–100	2	12	3	
mesoporous Fe–alumina films	21.70	2	30	3	1	4	
Nano UiO66–NH <sub>2</sub> MOF	32.36	1	—	6.50	24	5	
NH <sub>2</sub> -GO/ZnOZnFe <sub>2</sub> O <sub>4</sub> NMs	109.89	0.50	10–100	4	1.50	6	
HNT@CRC-8	140.87	0.40	100	2	50	7	
AO NPs	95.52	0.36 ± 0.00031	4.28–502.50	2	5		
PAA–AO NC beads	128.52	0.36 ± 0.00033	4.28–502.50	3	5		this work
PAA–AO–PEI NC beads	141.09	0.36 ± 0.00026	4.28–502.50	3	5		

$C_0$  stands for the initial concentrations; h stands for contact time in hours; ref. stands for references; NPs and NC stand for nanoparticles and nanocomposite, respectively.



**Figure S4.** Reusability tests of AO NPs and PAA–AO and PAA–AO–PEI NC beads for the adsorption of (A) Cr(VI) and (B) CR.

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