

Supporting Information (SI)

Guar Gum-Grafted Terpolymer Hydrogels for Ligand-Selective Individual and Synergistic Adsorption: Effect of Comonomer Composition

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EXPERIMENTAL SECTION

Methodology

MV and Hg(II) solutions of varying concentrations (i.e. 10–60 ppm) were prepared by exact dilution of 1000 ppm stock solutions. In the present study, 0.025 g of dry GGAMSAASPs were added to 50 mL buffered solutions of MV and Hg(II), with constant stirring at 300 rpm. The progress of adsorption was monitored by withdrawing supernatant solution after pre-determined time intervals, followed by measuring absorbance at $\lambda_{\max}$ using UV-vis spectrophotometer and atomic absorption spectrometer for MV and Hg(II), respectively. From the pre-calibrated equation, the dye concentration (C_t) was calculated to determine adsorption capacity (AC, q_t) (mg g^{-1}) using eq S1.

$$q_t = \frac{(C_0 - C_t)V}{m_s} \quad (\text{S1})$$

Here, C_0/C_t (ppm), V (mL) and m_s (g) are feed dye concentrations at $t = 0/t$, volume of adsorbate solutions and mass of GGAMSAASPs, respectively. However, equilibrium AC (q_e , mg g^{-1}) was obtained via replacing C_t by C_e in eq S1. The equilibrium data were fitted to the following adsorption isotherm models (eq S2–S4).

$$q_e = q_{\max} \frac{k_L C_e}{1 + k_L C_e} \quad (\text{S2})$$

$$q_e = k_F C_e^{1/n} \quad (\text{S3})$$

$$q_e = q_{\max} \frac{(k_S C_e)^\gamma}{1 + (k_S C_e)^\gamma} \quad (\text{S4})$$

Here, k_L , k_F , k_S are corresponding isotherm constants and q_{max} , n , γ are respective parameters of isotherm models.

RESULTS AND DISCUSSION

Swelling and pH reversibility studies of GGAMSAASPs

Swelling property of hydrogel is highly essential for studying hydrophilicity of the network that depends on the number of ionizable hydrophilic groups, such as $-\text{NH}_2$, $-\text{COOH}$, $-\text{OH}$, $-\text{CONH}_2$, and $-\text{CONH}-$ attached with the polymeric chains, along with the capillary effect and osmotic pressure. In the present case, swelling studies were carried out in buffer solutions of $\text{pH}_i = 3.0, 5.0, 7.0, 9.0$, and 12.0 at 30°C to find out the ESR. However, the maximum ESR was observed at $\text{pH}_i = 9$ for both the GGAMSAASPs. In fact, subsequent shrinking and poor swelling were observed for both the GGAMSAASPs in acidic pH_i of 3 and 5, owing to the lower population of $-\text{COO}^-$ resulting in lower electrostatic repulsion, which produced lesser space for water accommodation and hence, low ESR. In fact, at the swelled state, the coexistence of liquid and homogeneous gel phases has already been reported.¹ This infers that hydrogel is not completely dissolved in the solvent during swelling, yet retains the structural integrity even in the swollen state, owing to the presence of hydrophobic skeleton and certain degree of hydrophilic functional groups. These hydrophilic groups interact with water and cationic dyes/metal ions [i.e. M(II/III/VI)], but the overall hydrophobic structure restricts the dissolution of hydrogel in the polar solvent/solution(s). Thus, swelling does not result in the dissolution of GGAMSAASP network. In this context, the shrinking phenomenon of the swelled GGAMSAASP was reported by us, in the section of gel content measurement, in which

the known amount of dry GGAMSAASP was allowed to swell up to the equilibrium in double distilled water and the swelled GGAMSAASP was again dried to obtain the xerogel. This phenomenon clearly indicated the retention of structural integrity of GGAMSAASP during the time of swelling measurement. Moreover, the decrease in water absorbency at acidic pH_i could also be attributed to the protonation of $-\text{CONH}_2$ and $-\text{COO}^-$ leading to the decrease in H-bonding with water. At $\text{pH}_i = 9$, the occurrence of the maximum ESR for both the GGAMSAASPs (Figure 7c,d) could be explained via predominant population of $-\text{COO}^-$ resulting in the electrostatic repulsion, which led to macromolecular expansion and allowed large quantity of water to penetrate. However, at very high pH_i (i.e. $\text{pH}_i = 12$), rapid ionization of the available functional groups created significant amount of counter ion concentration inside the polymeric matrix, resulting in lower electrostatic repulsion and hence, lower ESR. Furthermore, the formation of hydration sheath around the polymeric network by aqua-ions at higher pH_i might cause significant reduction in the degree of ionization and ESR. Moreover, GGAMSAASP18 showed reduced swelling with the increase in ionic strength of the solution (Figure S6).

The pH reversibility of GGAMSAASPs was ascertained via repeating swelling/deswelling studies at low/high pH_i (3/10) (Figure 7a). A 0.01 g of xerogel was first immersed into $\text{pH}_i = 3$ for 1 h, followed by immersing into solution of $\text{pH}_i = 10$ for another 1 h, and the same procedure was continued for several cycles until GGAMSAASPs remained stable. However, after 3 complete cycles of immersion, both the AMSAASPs became fragile, whereas the GGAMSAASPs exhibited enough stability to withstand up to 5 complete cycles. In fact, both the GGAMSAASPs exhibited high pH reversibility and hence, $\text{pH}_i > \text{pH}_{\text{PZC}}$ was chosen for adsorption.

Calculation of % gel content (%GC), % graft ratio (%GR), and pH_{PZC} of GGAMSAASPs

The %GC of GGAMSAASPs were estimated by a method reported elsewhere using eq S5.^{S34}

$$\%GC = \frac{W_d}{W_i} \times 100 \quad (\text{S5})$$

Accurately weighed air-dried GGAMSAASPs were further dried in a vacuum oven at 50 °C to obtain a constant weight (W_i). Then, it was dispersed in distilled water for 72 h with occasional stirring to fully swell them for elimination of water soluble components from the network. The water insoluble GGAMSAASPs were then dried in vacuum oven until a constant weight (W_d) was obtained. However, %GC of GGAMSAASP14/18 were found to be 75.64/92.37 %. The %GR of GGAMSAASPs were calculated by employing eq S6.^{S34}

$$\%GR = \frac{W_2}{W_1} \times 100 \quad (\text{S6})$$

Here, W_2 and W_1 represent masses of GG and GGAMSAASPs, respectively. The %GR of the used GGAMSAASP18/14 were obtained to be 2.62/3.20 %.

The pH_{PZC} of both the GGAMSAASPs were estimated by a method reported elsewhere.^{S34} In this context, 0.05 g of xerogel was taken in 50 mL buffer solutions of different pH_i within 2–10. After 72 h of immersion, final pH (pH_f) of all the solutions were estimated. The difference of these pH_f and pH_i was plotted against pH_i to find the pH_{PZC} . However, pH_{PZC} were found to be 6.07 and 5.85, for GGAMSAASP18 and GGAMSAASP14, respectively (Figure 5b).

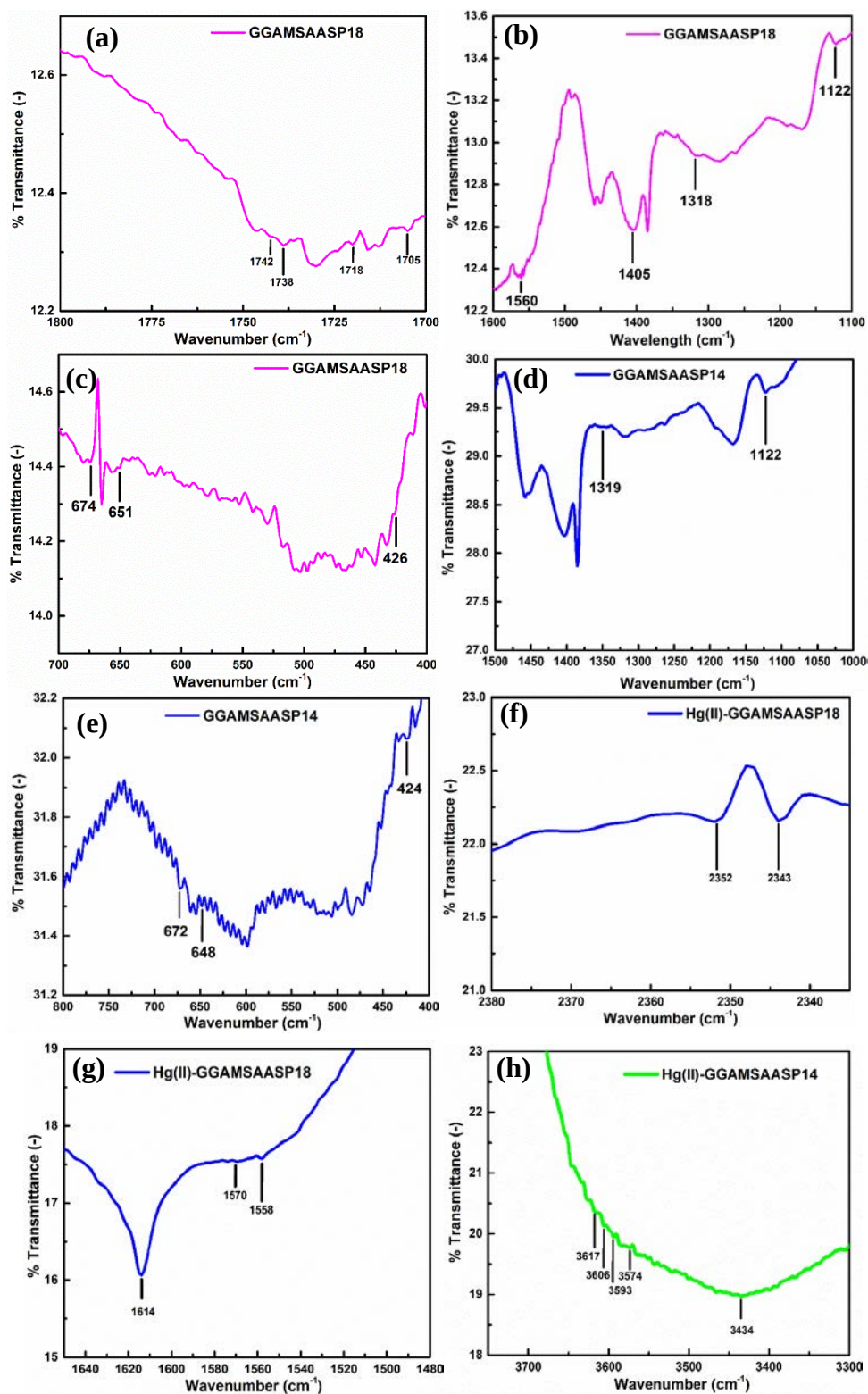
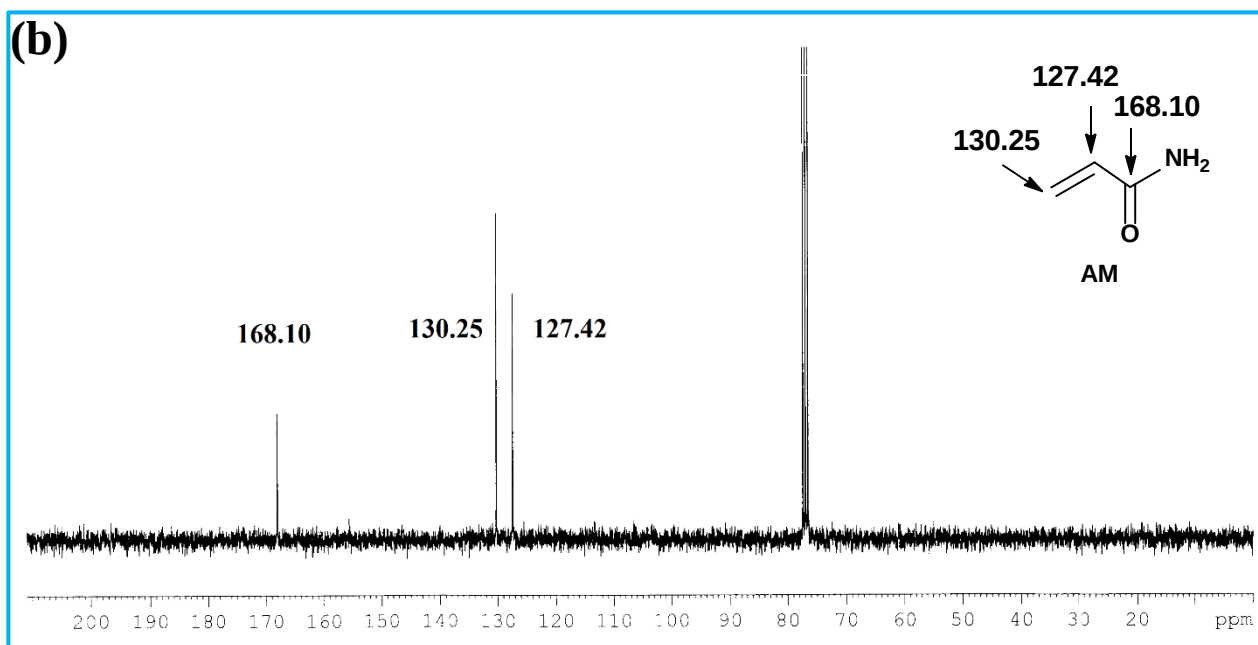
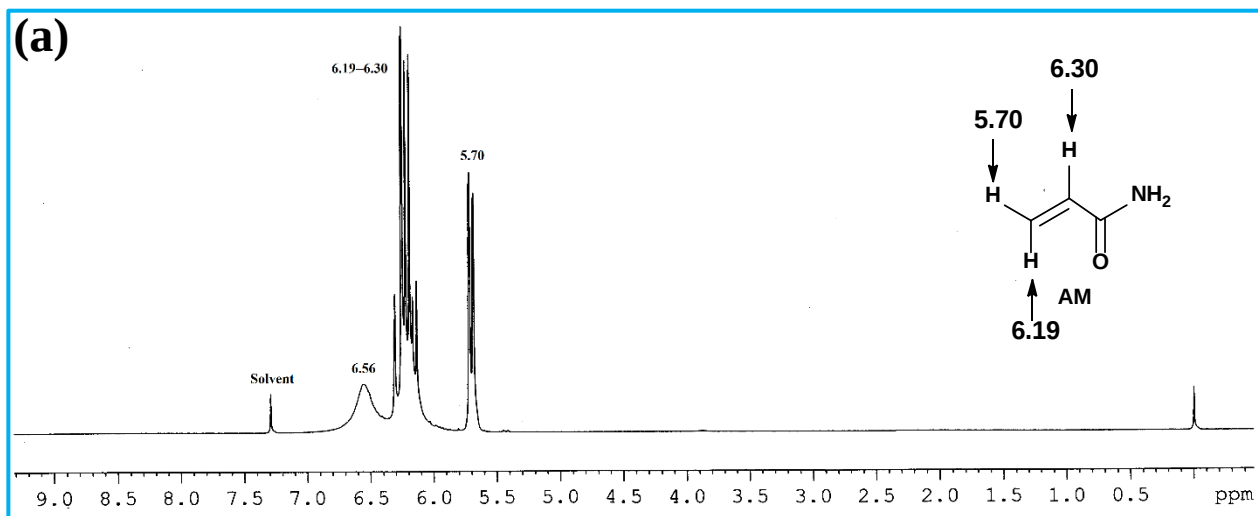
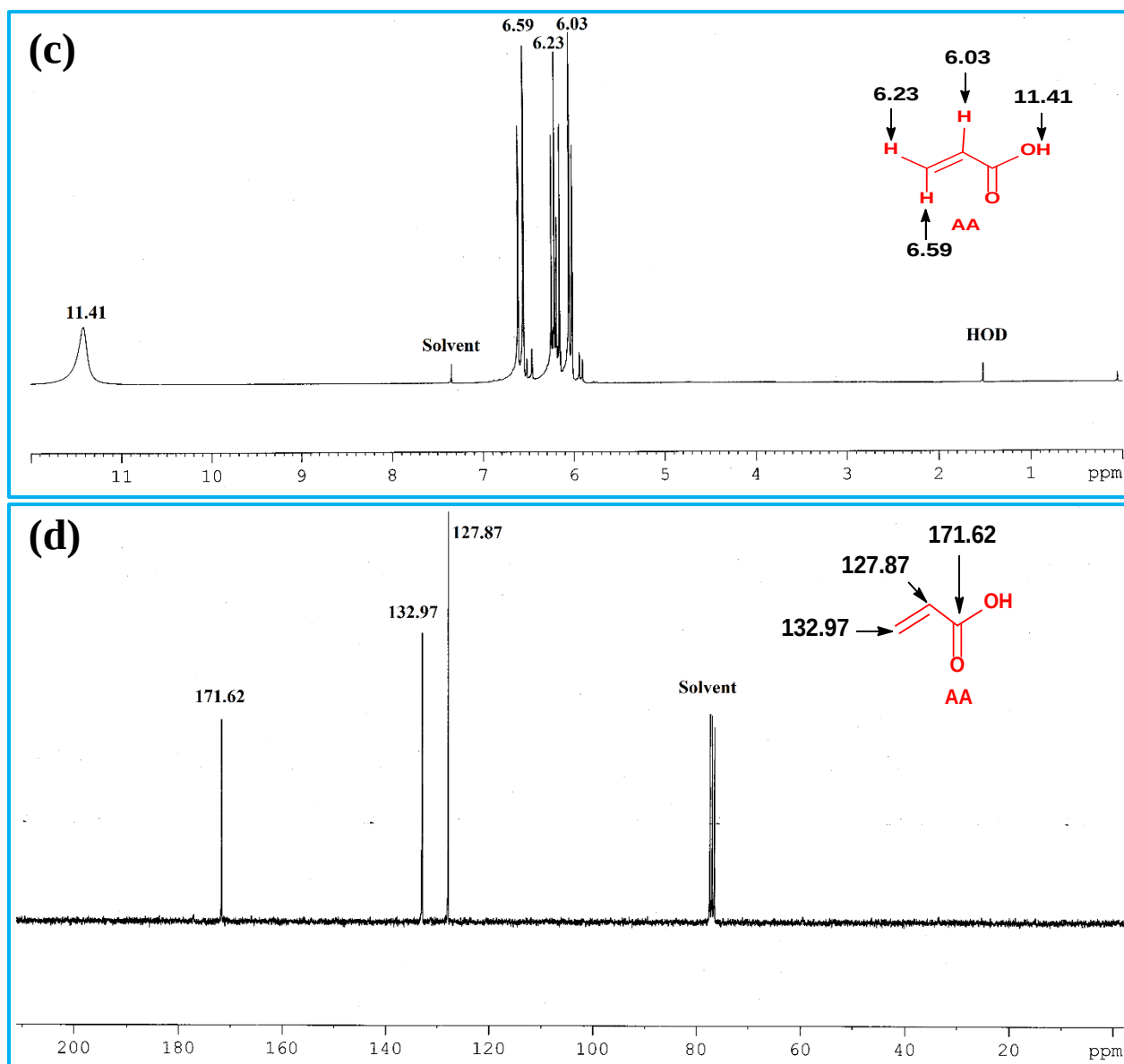
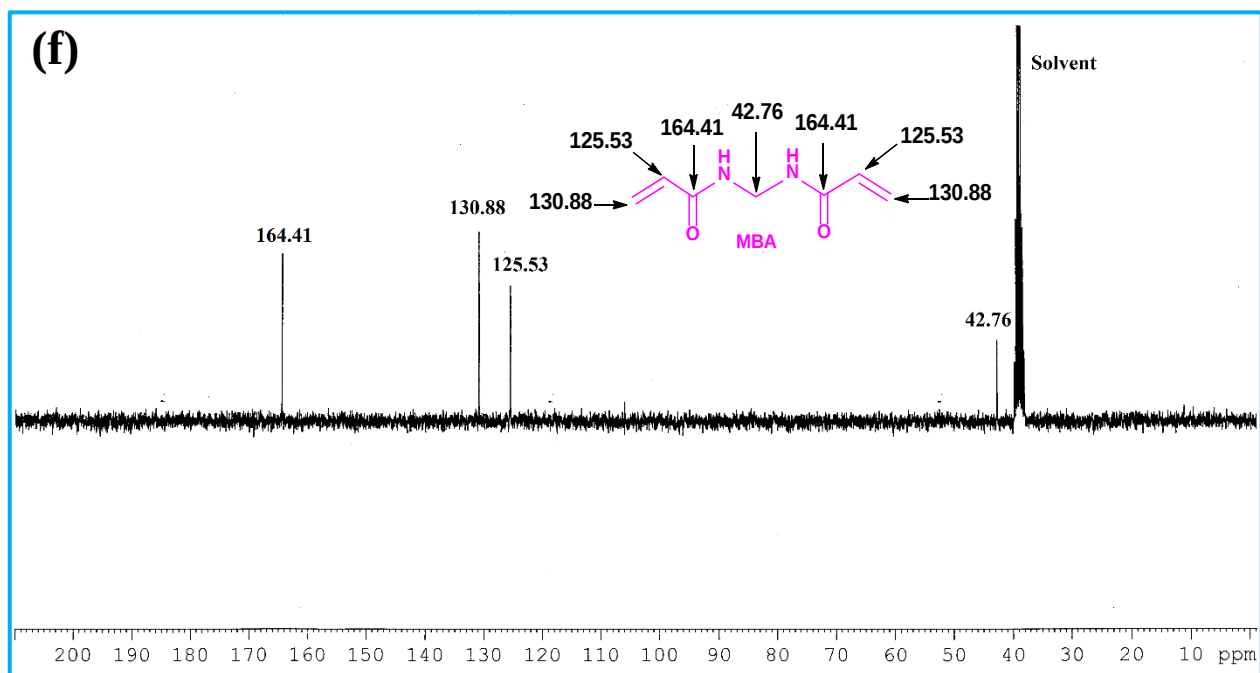
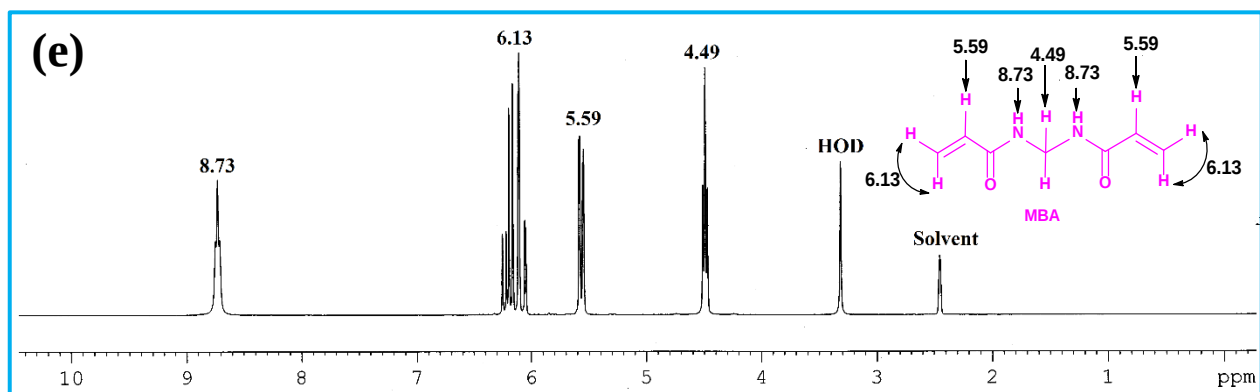


Figure S1. FTIR of (a/b/c) GGAMSAASP18, (d/e) GGAMSAASP14, (f/g) Hg(II)-GGAMSAASP18 and (h) Hg(II)-GGAMSAASP14







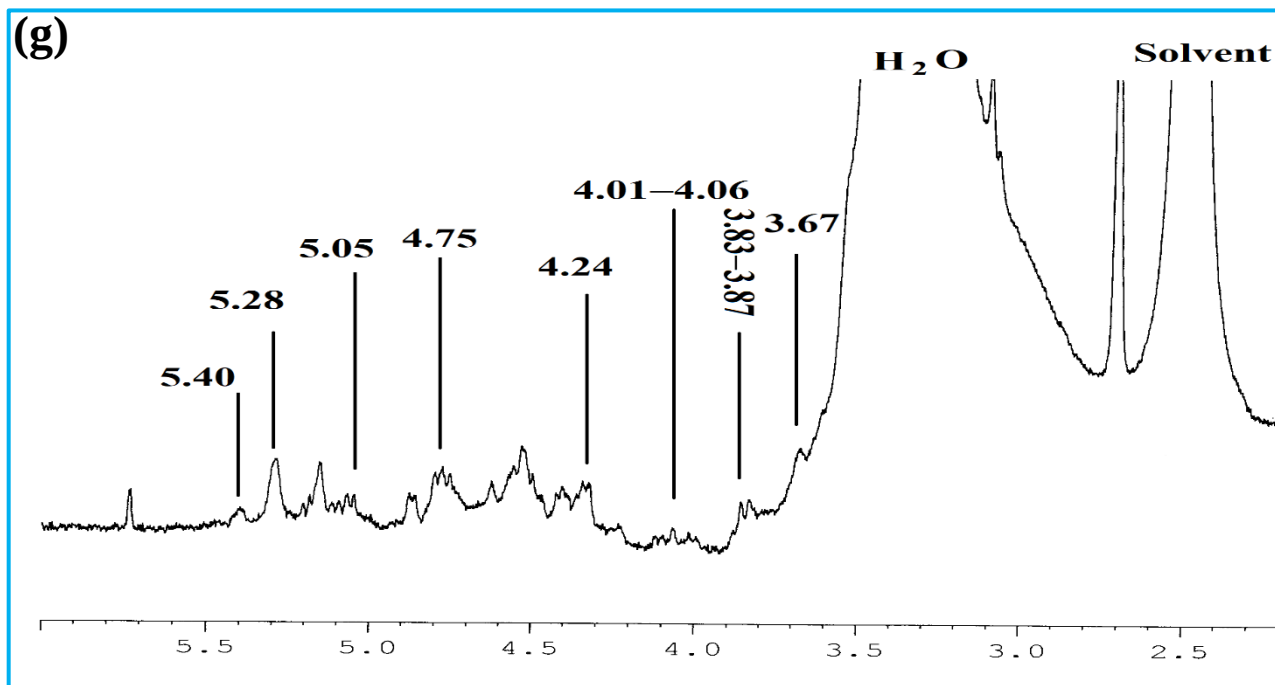


Figure S2. ^1H -NMR of (a) AM, (c) AA, (e) MBA and (g) GG and ^{13}C -NMR of (b) AM, (d) AA and (f) MBA

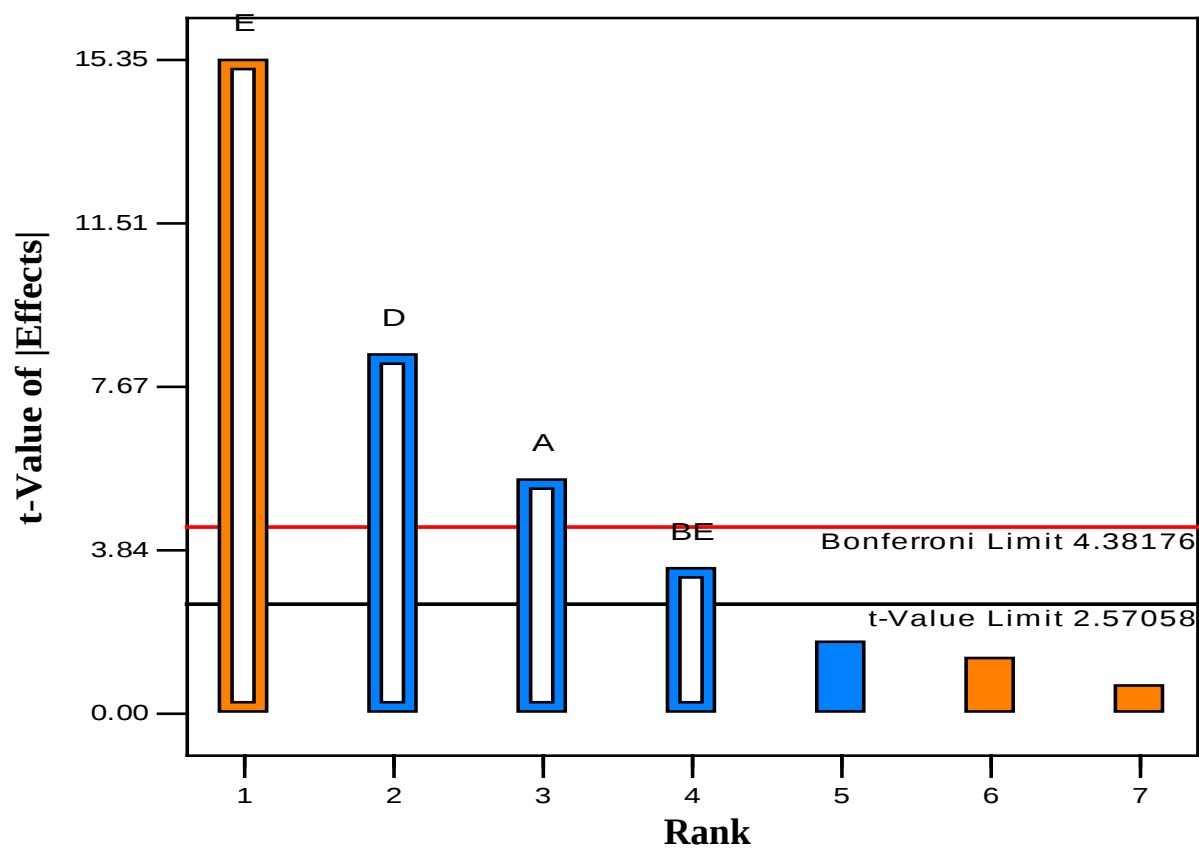


Figure S3. Pareto chart for screening of synthetic parameters

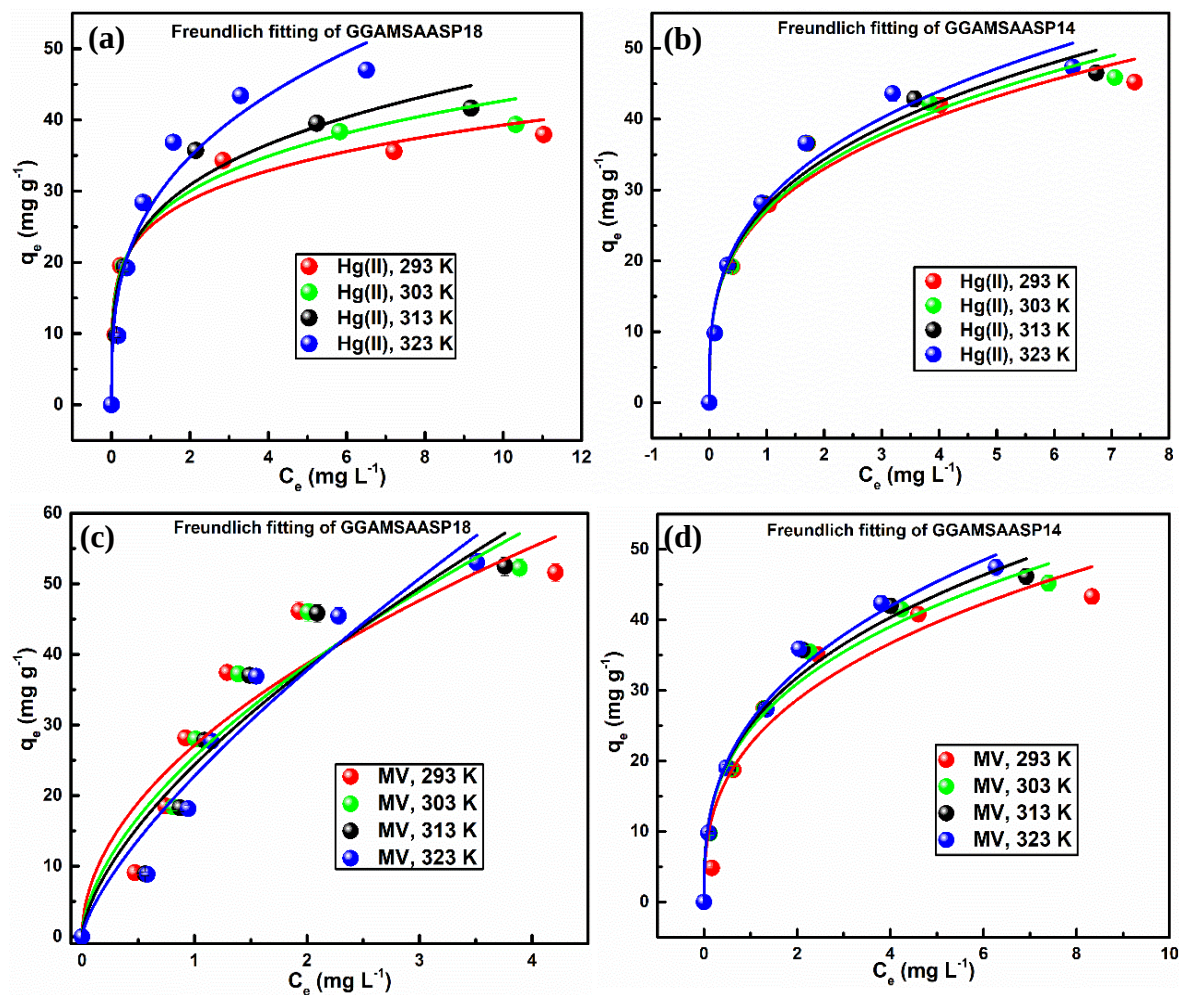


Figure S4. Freundlich fitting for (a/b) Hg(II)-GGAMSAASP18/14 and (c/d) MV-GGAMSAASP18/14

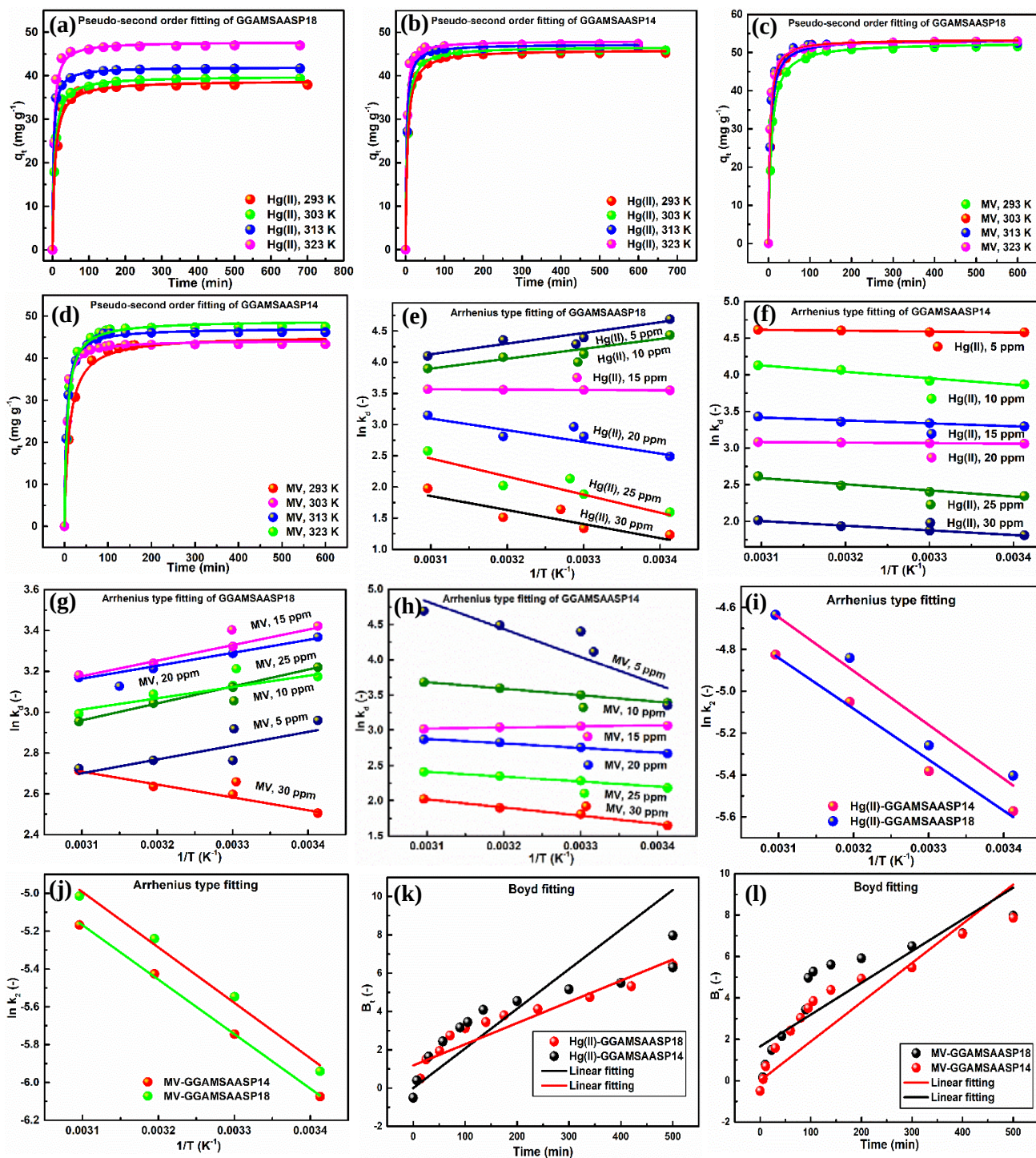


Figure S5. Pseudo-second order kinetics plots for (a/b and c/d) Hg(II)-GGAMSAASP18/14 and MV-GGAMSAASP18/14; $\ln k_d$ vs. $1/T$ for (e/f and g/h) Hg(II)-GGAMSAASP18/14 and MV-GGAMSAASP18/14; $\ln k_2$ vs. $1/T$ plots for (i and j) Hg(II)-GGAMSAASP18/14 and MV-GGAMSAASP18/14; Boyd fitting for (k and l) Hg(II)-GGAMSAASP18/14 and MV-GGAMSAASP18/14

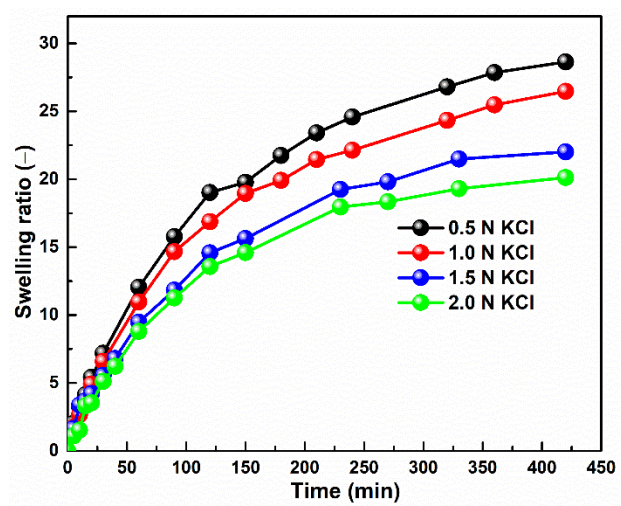


Figure S6. ESR of GGAMSAASP18 in various ionic strengths of solutions

Table S1. Center Composite Design of Experiment

run no.	amount of AM (wt %)	total amount crosslinker (wt %)	pH _i (-)	ESR (-)
1	6.25	1.00	4.00	5.50
2	25.00	1.00	4.00	5.10
3	6.25	5.00	4.00	3.30
4	25.00	5.00	4.00	2.90
5	6.25	1.00	12.00	9.22
6	25.00	1.00	12.00	7.68
7	6.25	5.00	12.00	5.56
8	25.00	5.00	12.00	2.21
9	0.00	3.00	8.00	10.80
10	31.39	3.00	8.00	8.32
11	15.62	0.00	8.00	7.90
12	15.62	6.36	8.00	3.00
13	15.62	3.00	1.27	2.33
14	15.62	3.00	13.00	10.34
15	15.62	3.00	8.00	18.40
16	15.62	3.00	8.00	18.40
17	15.62	3.00	8.00	18.40
18	15.62	3.00	8.00	18.40
19	15.62	3.00	8.00	18.40
20	6.25	3.00	8.00	18.40

Table S2. ANOVA Statistics of CCD

source	sum of squares	degrees of freedom	mean square	<i>F</i> value	<i>p</i> -value
model	759.70	9	84.41	213.80	< 0.0001*
amount of AM (A)	6.74	1	6.74	17.07	0.0020*
amount of crosslinker (B)	21.29	1	21.29	53.92	< 0.0001*
pH _i (C)	12.78	1	12.78	32.37	0.0002*
AB	0.41	1	0.41	1.037	0.3325
AC	2.09	1	2.09	5.30	0.0442*
BC	2.80	1	2.80	7.08	0.0238*
A ²	141.64	1	141.64	358.76	< 0.0001*
B ²	307.61	1	307.61	779.11	< 0.0001*
C ²	283.46	1	283.46	717.95	< 0.0001*
residual	3.95	10	0.39		
lack of fit	3.95	5	0.79		
pure error	0.00	5	0.00		
cor. total		19			
std. dev.	0.63		<i>R</i> ²	0.9948	
mean	9.73		adj. <i>R</i> ²	0.9901	
CV %	6.46		pred. <i>R</i> ²	0.9597	
PRESS	30.76		adeq. precision	36.6606	

* significant

Table S3. Comparative Table

name of adsorbate	name of adsorbent	adsorption capacity (mg g ⁻¹) /pH _i /C ₀ (ppm)/temperature (K)	ref.
Hg(II)	Chitosan derivative adsorbent	9.02/3.0/60/298	S1
	RGO ^a -MnO ₂	9.50/-/1/303	S2
	RGO ^a -Ag	9.53/-/1/303	S2
	APT ^b	13.20/5.0/3800/303	S3
	<i>Hardwickia binata</i> bark	13.50/6.0/400/298	S4
	Natural chitosan spheres	13.50±0.40/6.0/38–375/298	S5
	Mesoporous silica-coated magnetic particles	14.00/2.0/10–60/-	S6
	poly(AAm-co-AAc) ^c	15.50/2.5/100/288	S7
	Ti(IV) ^d	17.20/6.0/20/293–323	S8
	SMs ^e	20.00/7.5/100–900/303	S9
	GMA-MMA-DVB ^f	20.06/7.0/15/298	S10
	Chemically treated sawdust (<i>Acacia arabica</i>)	20.62/6.0/3/-	S11
	Multifunctional mesoporous material	21.05/-/1000/-	S12
	CTS-PVA ^g	24.98/5.5/50/303	S13
	Ca-alginate beads	28.90±0.70/6.0/200/298	S14
	Poly(MMA-MAGA) ^h	29.90/2.0–6.0/100/293	S15
	Epichlorohydrin-crosslinked chitosan membranes	30.30/6.0/38–375/298	S16
	Glutaraldehyde-crosslinked chitosan spheres	31.10±0.30/6.0/38–375/298	S5
	BTESPT-SMs ⁱ	37.00/7.5/100–900/303	S9
	Cellulose–Lysine–Schiff Bases	50.60/4.4/100/303	S17
	TCPF ^j	52.63/6.0/50/301	S18
	4-aminoantipyrine immobilized bentonite	52.90/4.0/1/298	S19
	CNTs/Fe ₃ O ₄ ^k	65.52/6.5/50/298	S20
	GGAMSAASP18^l	40.95/7.0/5–30/303	TS
	GGAMSAASP14^m	49.12/7.0/5–30/303	TS
MV	CPSA4 ⁿ	2.09/7.0/2/298	S21
	Semi-IPN of starch and copolymer of AM ^o and HEMA ^p	2.47/7.0/2.5/303	S22
	Bagasse fly ash	3.712/9.0/10/303	S23
	Poly(VP-co-MA) ^q	4.22/7.0/500/298	S24
	Soya ash	5.76/9.0/25/303	S25
	Poly(AM-co-AA) ^r	6.38/7.0/50/298	S26
	Orange peel	11.50/5.3/100/303	S27
	Banana peel	12.20/5.3/100/303	S27
	Mansonia (<i>Mansonia altissima</i>) wood sawdust	16.11/10.0/120/299	S28
	MSWI bottom ash ^s	19.58/8.0/24/303	S29
	Mansonia wood sawdust	20.20/10.0/30–120/309	S30
	Cereal chaff	20.30/11.0/30/293	S31
	PAAC ^t	49.96/9.2/25/295	S32
	Halloysite nanotube-Fe ₃ O ₄ composite	64.40/9.0/150/298	S33
	IPNS ^u	21.68/10.0/5–30/303	S34

^aReduced graphene oxide, ^battapulgit, ^cpoly(acrylic acid/acrylamide), ^dTi(IV) iodovanadate cation exchanger, ^esilica microspheres, ^fmethyl methacrylate-glycidyl methacrylate-divinylbenzene terpolymer beads, ^gchitosan-poly(vinyl alcohol), ^hpoly(methyl methacrylate-methacryloylamidoglutamic acid), ⁱbis(triethoxysilylpropyl) tetrasulfide silica microspheres, ^jthiocarbohydrazide cross-linked chitosan-poly(vinyl alcohol) framework, ^kcarbon nanotube/magnetite nanocomposites, ^lguar gum-*g*-(acrylamide-*co*-sodium acrylate-*co*-acrylamidosodium propanoate)18, ^mguar gum-*g*-(acrylamide-*co*-sodium acrylate-*co*-acrylamidosodium propanoate)14, ⁿIPN of poly(acrylic acid-*co*-acrylamide) and sodium alginate, ^oacrylamide, ^phydroxyethyl methacrylate, ^qpoly(N-vinylpyrrolidone-*co*-methacrylic acid), ^rpoly(acrylamide-*co*-acrylic acid), ^smunicipal solid waste incinerator, ^t*phragmites australis* activated carbon, and ^uinterpenetrating polymer network superadsorbent

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