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 λ_{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.

\[ q_t = \frac{(C_0 - C_t)V}{m_s} \]  

(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).

\[ q_e = q_{max} \frac{k_L C_e}{1 + k_L C_e} \]  

(S2)

\[ q_e = k_F C_e^{1/n} \]  

(S3)

\[ q_e = q_{max} \frac{(k_L C_e)^y}{1 + (k_L C_e)^y} \]  

(S4)
Here, $k_L$, $k_F$, $k_S$ are corresponding isotherm constants and $q_{\text{max}}$, $n$, $\gamma$ 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 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 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\textsubscript{i} could also be attributed to the protonation of –CONH\textsubscript{2} and –COO\textsuperscript{−} leading to the decrease in H-bonding with water. At pH\textsubscript{i} = 9, the occurrence of the maximum ESR for both the GGAMSAASPs (Figure 7c,d) could be explained via predominant population of –COO\textsuperscript{−} resulting in the electrostatic repulsion, which led to macromolecular expansion and allowed large quantity of water to penetrate. However, at very high pH\textsubscript{i} (i.e. pH\textsubscript{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\textsubscript{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\textsubscript{i} (3/10) (Figure 7a). A 0.01 g of xerogel was first immersed into pH\textsubscript{i} = 3 for 1 h, followed by immersing into solution of pH\textsubscript{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, pH\textsubscript{i} > pH\textsubscript{PZC} was chosen for adsorption.
Calculation of % gel content (%GC), % graft ratio (%GR), and pH\textsubscript{PZC} of GGAMSAASPs

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

\[
\%GC = \frac{W_2}{W_i} \times 100
\]  

(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 GGAMSAASP\textsubscript{14/18} were found to be 75.64/92.37 %. The %GR of GGAMSAASPs were calculated by employing eq S6. \cite{S34}

\[
\%GR = \frac{W_2}{W_1} \times 100
\]  

(S6)

Here, \(W_2\) and \(W_1\) represent masses of GG and GGAMSAASPs, respectively. The %GR of the used GGAMSAASP\textsubscript{18/14} were obtained to be 2.62/3.20 %.

The pH\textsubscript{PZC} of both the GGAMSAASPs were estimated by a method reported elsewhere. \cite{S34} In this context, 0.05 g of xerogel was taken in 50 mL buffer solutions of different pH\textsubscript{i} within 2–10. After 72 h of immersion, final pH (pH\textsubscript{f}) of all the solutions were estimated. The difference of these pH\textsubscript{f} and pH\textsubscript{i} was plotted against pH\textsubscript{i} to find the pH\textsubscript{PZC}. However, pH\textsubscript{PZC} were found to be 6.07 and 5.85, for GGAMSAASP\textsubscript{18} and GGAMSAASP\textsubscript{14}, 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. $^1$H-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. Pseudosecond 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 | F value | p-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, (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*   |
| A²                                  | 141.64         | 1                  | 141.64      | 358.76  | < 0.0001* |
| B²                                  | 307.61         | 1                  | 307.61      | 779.11  | < 0.0001* |
| C²                                  | 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                          |                |                    |             |         |           |
| std. dev.                           | 0.63           |                    | R²          | 0.9948  |           |
| mean                                | 9.73           |                    | adj. R²     | 0.9901  |           |
| CV %                                | 6.46           |                    | pred. R²   | 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) | ref. |
|-------------------|-------------------|-------------------------------|------|
| **Hg(II)**        | Chitosan derivative adsorbent | 9.02/3.0/60/298 | S1   |
|                   | RGO^a-MnO₂         | 9.50/−/1/303                 | S2   |
|                   | RGO^a-Ag           | 9.53/−/1/303                 | S2   |
|                   | APT^b              | 13.20/5.0/3800/303           | S3   |
| **Hardwickia binata 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 (Acacia arabica)** |               | 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/Fe₃O₄^k**  |                   | 65.52/6.5/50/298            | S20  |
| **GGAMSAAASP18^l** |                | 40.95/7.0/5−30/303         | TS   |
| **GGAMSAAASP14^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  |
| **Mansonlia (Mansonia altissima) wood sawdust** | | 16.11/10.0/120/299 | S28  |
| **MSWI bottom ash^s** |              | 19.58/8.0/24/303           | S29  |
| **Mansonlia 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-Fe₃O₄ composite** |               | 64.40/9.0/150/298          | S33  |
| **IPNS^u**        |                   | 21.68/10.0/5−30/303        | S34  |
GGAMSAASP18
53.28/9.0/5–30/303
TS
GGAMSAASP14
50.29/9.0/5–30/303
TS

*aReduced graphene oxide, *battapulgite, *c(poly(acrylic acid/acrylamide), *d(Ti(IV) iodovanadate cation exchanger,*e)silica microspheres, *fmethyl methacrylate-glycidyl methacrylate-divinylbenzene terpolymer beads, *gchitosan–poly(vinyl alcohol), *hpoly(methyl methacrylate–methacryloyloxyethylidoglutamic acid), *jis(triethoxysilylpropyl) tetrasulfide silica microspheres, *kthinocarbohydrazide cross-linked chitosan-poly(vinyl alcohol) framework, *lcarbon nanotube/magnetite nanocomposites, *mguar gum-(acrylamide-‐co-‐sodium acrylate-‐co-‐acrylamidosodium propanoate)18, *nguar gum-‐(acrylamide-‐co-‐sodium acrylate-‐co-‐acrylamidosodium propanoate)14, *nIPN of poly(acrylic acid-‐co-‐acrylamide) and sodium alginate, *apoly(acrylic acid-‐co-‐acrylamide), *bpoly(acrylamide-co-acrylic acid), *cmunicipal solid waste incinerator, *dphragmites australis activated carbon, and *einterpenetrating polymer network superadsorbent

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