Supporting Information

Designed synthesis of sulfide-rich bimetallic-assembled graphene oxide sheets as flexible materials and *self-tuning* adsorption *cum* oxidation mechanisms of arsenic from waters

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Analytical methods

The functional groups of the as-synthesized materials before and after adsorption of arsenite and arsenate were characterized by FTIR (PerkinElmer, Frontier) under transmission mode. Powdered XRD (PXRD) patterns were carried out on Ultima IV diffractometer (RIGAKU, D/Max-2500). Raman spectra were done using Renishaw, inVia reflex model. The morphology of the as-synthesized adsorbents and after treatment with arsenite/arsenate was investigated by the field emission scanning electron microscope (FE-SEM, Hitachi, SU-8220). The pH of the suspension was adjusted to analyze the Zeta potentials by a ZETA SIZER NANO-Z (Malvern, Kobe, Japan). X-ray photoelectron spectroscopy (XPS) measurements were conducted on an ESCA 5800 (ULVAC-PHI, Inc, Kanagawa, Japan) using a monochromated Al Kα X-ray source at 200 W for Zr 3d, As 3d, C 1s and O 1s, and Casa XPS software (Version 2.3.12.8) was used to analyze. E_B[C 1s] = 284.6 eV for adventitious carbon was used to calibrate the binding energies (E_B). The transmission electron microscopy (TEM) pattern was performed on a JEOL, JEM-2100F electron microscope (Japan) operating at 200 kV. The powder samples were added in ethanol in an ultrasonic bath for 5 min and deposited on a copper grid covered with perforated carbon film.

Kinetic models

The linear forms of Lagergren pseudo-first-order and Ho's pseudo-second-order kinetic models are as follows:

$$\log \left(q_e - q_t\right) = \log q_e - \frac{k_1}{2.303}t$$

(1)

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \tag{2}$$

where k_1 and k_2 are rate constants, and q_e is the adsorption capacity (mmol/g) at the equilibrium. A straight line in the plot of t/q_t vs t, and the values of its intercept and slope would suggest a proper application of the pseudo-second-order kinetic model to represent the arsenite and arsenate adsorption on the surface of the adsorbent under the given experimental conditions.

Langmuir isotherm

The linear form of Langmuir adsorption isotherm is expressed by:

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \frac{C_e}{q_m} \tag{3}$$

where C_e (mmol/L) is the equilibrium concentration of arsenate and arsenate after adsorption; q_e (mmol/g) is the adsorption capacity at equilibrium; q_m is the calculated maximum adsorption capacity (mmol/g); and b (L/mmol) is the Langmuir constant.

Freundlich isotherm

The linear form of Freundlich adsorption isotherm is expressed by:

$$\log q_e = \log K_F + 1/n \log C_e \tag{4}$$

where K_F is the Freundlich adsorption constant; and $1/n$ denotes the interaction of adsorbate and adsorbent.

Table S1. Kinetics parameters calculated for arsenite and arsenate adsorption onto CLS50%@GO nanohybrid.

Adsorbent	Pseudo-first-order				
	Adsorbate	q_{exp} (mmol/g)	q_{cal} (mmol/g)	k_1 (1/min)	R^2
CLS50%@GO	AsO_4^{3-}	0.9815	0.7124	1.0562	0.8437
	AsO_3^{3-}	0.5886	0.4586	0.4198	0.7456

| Adsorbent	Pseudo-second-order				
	Adsorbate	q_{exp} (mmol/g)	q_{cal} (mmol/g)	k_2 (g/mmol min)	R^2
CLS50%@GO	AsO_4^{3-}	0.9815	0.9885	0.0923	0.9998
	AsO_3^{3-}	0.5886	0.6167	0.0270	0.9945
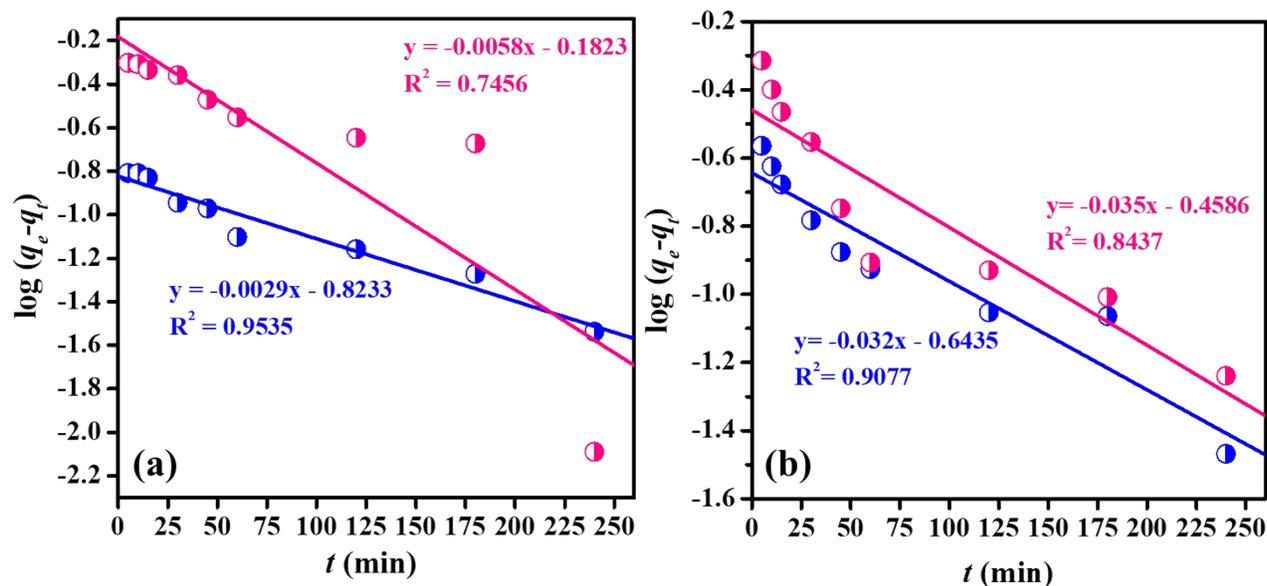
Fig. S1. Pseudo-first-order kinetic model for adsorption of (a) arsenite and (b) arsenate on CLS (blue color) and CLS50%@GO (pink color). Experimental conditions: $[AsO_3^{3-}]_0$ (pH) = 0.9 mM (9.2); $[AsO_4^{3-}]_0$ (pH) = 1.1 mM (5.5); dose = 1 g/L; reaction time = 24 h; agitation = 100 rpm; and temperature = 25 ºC.
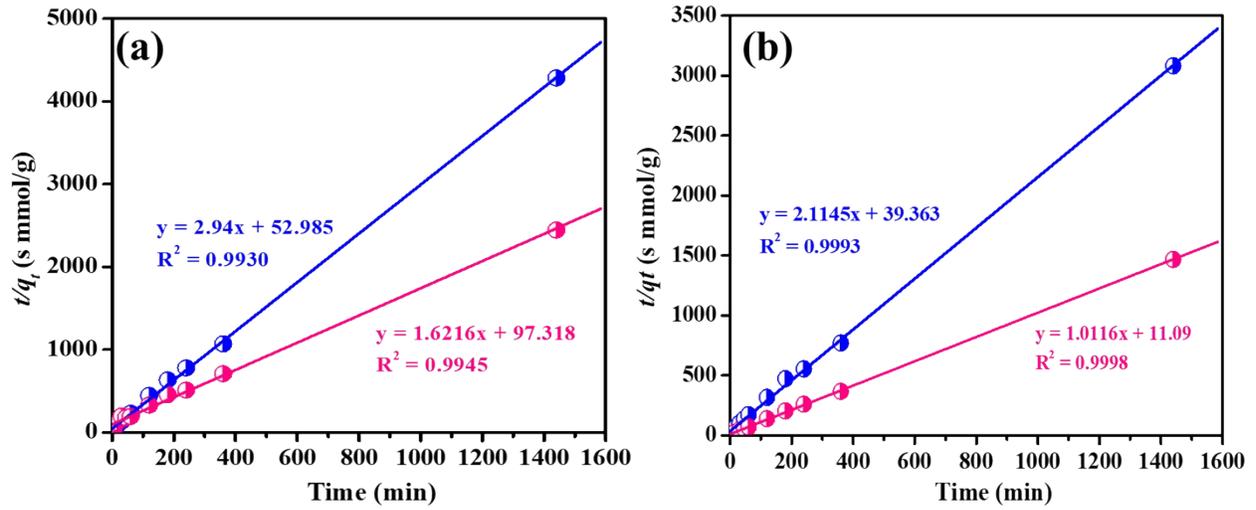
Fig. S2. Pseudo-second-order kinetic model for adsorption of (a) arsenite and (b) arsenate on CLS (blue color) and CLS50%@GO (pink color). Experimental conditions: $[AsO_3^{3-}]_0$ (pH) = 0.9 mM (9.2); $[AsO_4^{3-}]_0$ (pH) = 1.1 mM (5.5); dose = 1 g/L; reaction time = 24 h; agitation = 100 rpm; and temperature = 25 ºC.
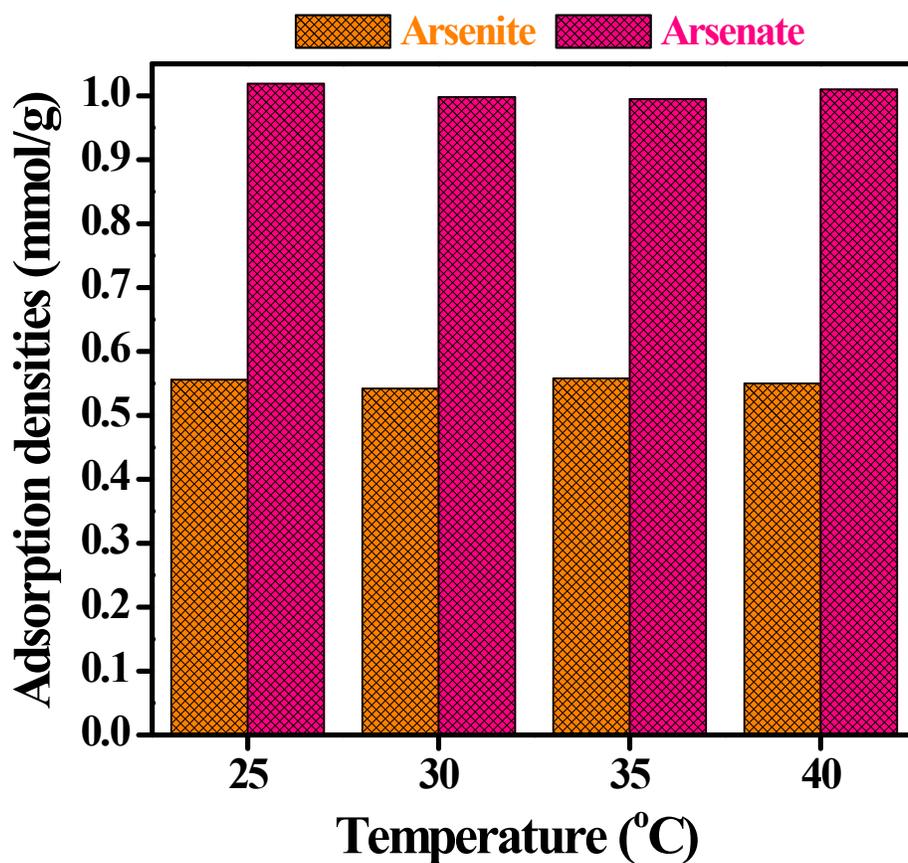
Fig. S3. Effect of temperature for the adsorption of arsenite and arsenate on CLS50%@GO. Experimental conditions: $[AsO_3^{3-}]_0$ (pH) = 0.9 mM (9.2); $[AsO_4^{3-}]_0$ (pH) = 1.1 mM (5.5); dose = 1 g/L; reaction time = 24 h; agitation = 100 rpm; and temperature = 25, 30, 35, and 40 ºC.
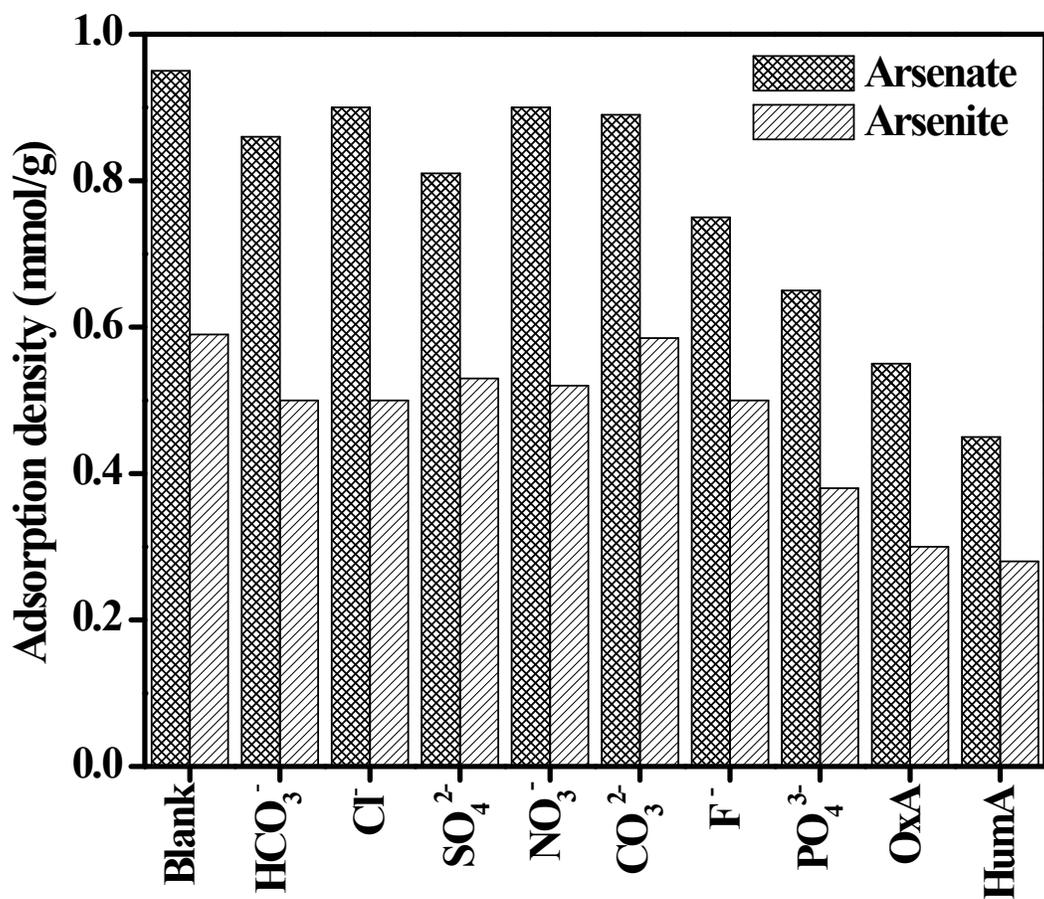
Fig. S4. Effect of coexisting anions. Experimental conditions: $[AsO_3^{3-}]_0 = 0.9$ mM; $[AsO_4^{3-}]_0 = 1.1$ mM; dose = 1 g/L; reaction time = 24 h; agitation = 100 rpm; and temperature = 25 ℃.
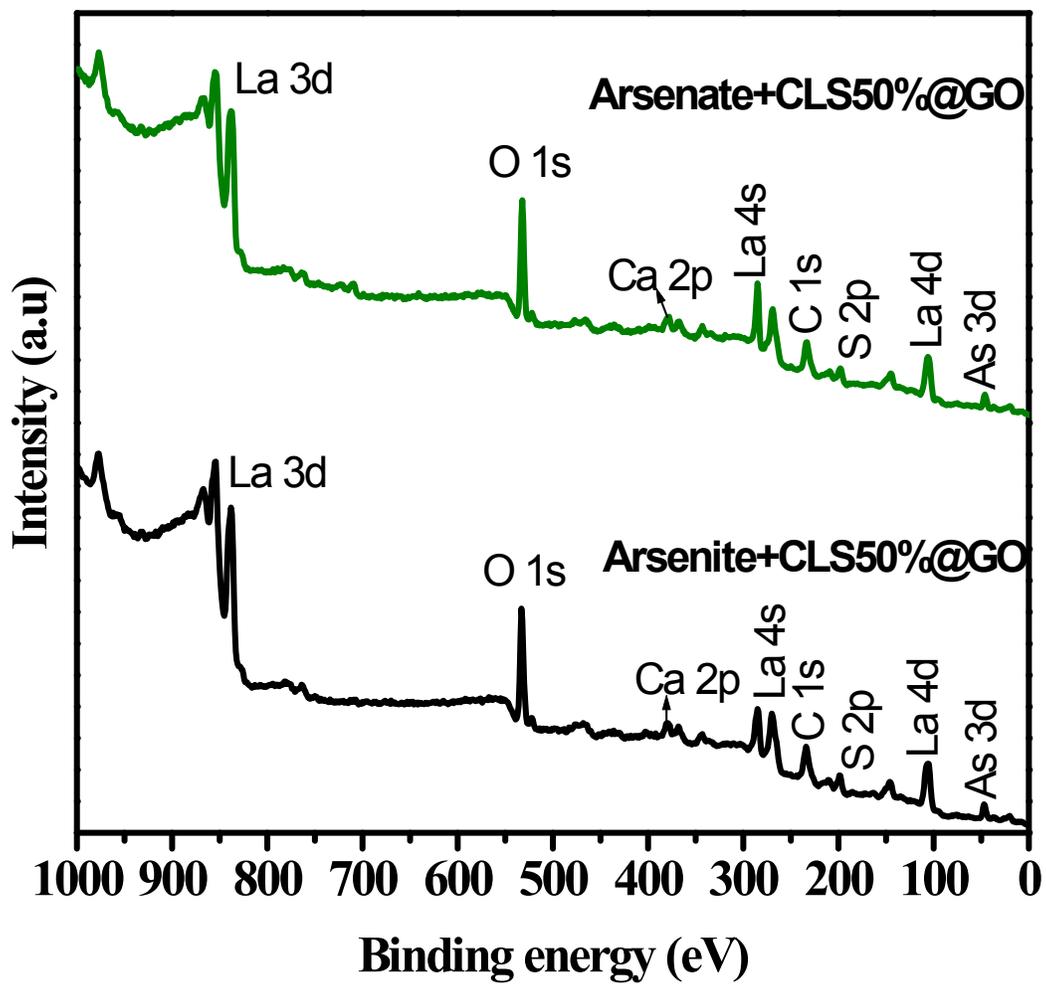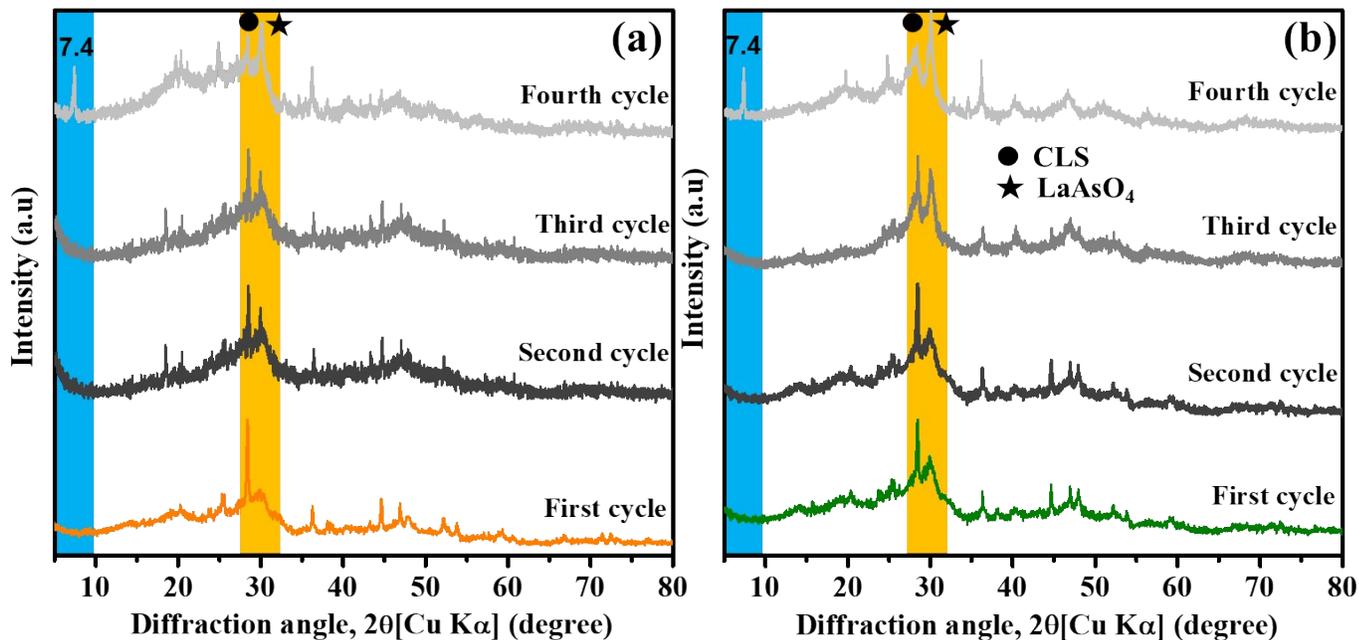
Fig. S5. Wide XPS spectra of arsenite and arsenate sorbed CLS50%@GO nanohybrids.

Fig. **S6.** PXRD patterns of the CLS50%@GO after each regeneration cycles of (a) arsenite and (b) arsenate using 0.1 M HNO_3 as eluent.