Supporting information

A Selective and Practical Graphene-based Arsenite Sensor at 10 ppb

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S1. Experimental methods

(i) Synthesis of GO

GO was synthesized from graphite powder based on the modified Hummer's method (Step1 of Figure S1). Briefly, graphite powder (2 g) was oxidized in a hot solution (100°C) of concentrated H_2SO_4 (25 mL), $K_2S_2O_8$ (4 g), and P_2O_5 (4 g). The resulting dark blue mixture was allowed to cool to room temperature over a period of 6 h. The mixture was diluted to 200 mL with DI water and the solution was filtered. Finally, the filtrated product was dried overnight at 60°C in a hot air oven. As synthesized pre-oxidized graphite powder (2 g) was further added to 92 mL of cold H_2SO_4 (0°C). $KMnO_4$ (12 g) was gradually added to the mixture under continuous stirring in ice-bath. After 15 min, $NaNO_3$ (2 g) was added to the mixture. The solution was further stirred for 2 h at 35 °C and distilled water (200 mL) was added dropwise during stirring. The reaction was stopped after simultaneous addition of a mixture of 300 mL distilled water and 10 mL of H_2O_2 (30 %) to the solution. The final product was washed sequentially with different solvents; at first, with diluted HCl (1:10) and then with water, and at last, the product was suspended in distilled water. The brown dispersion was dialyzed extensively to remove residual metal ions and acids. Finally, the dispersion was exfoliated via ultrasonication (300 W) for 2 h and unexfoliated graphite oxide was removed by centrifugation (12000 rpm for 20 min using Centrifuge KUBOTA (Tokyo, Japan)).

(ii) ERGO fabrication on Au coated test strips

At first, flexible and patterned Au strips, on which ERGO was fabricated, was pre-treated with 3 mM of sodium 2-mercaptoethanesulfonate (MESA, $HSCH_2CH_2SO_3Na$). Au strips were dipped in MESA for 72 h to create self-assembled monolayer of thiols on the Au surface. The aim of thiol pre-treatment was to deposit a self-assembled monolayer (SAM) on the Au surface (Step 2 of Figure S1). Test strips were rinsed to remove excess thiols from the Au surface, and dried under N_2 gas. To check the electrochemical activity of thiol modified Au surface, we measured cyclic voltammetry (CV) of the test strips (with and without thiol treatment) with a

solution mixture of 1 mM potassium ferricyanide and 100 mM KCl. The resulting voltammogram is shown in Figure S1 (Step 3), where the potential difference of redox peak (ΔE_p) of thiol treated sample was 60-80 mV, which ensures a one electron transfer process at the interface between the SAM modified Au strip and the electrolyte. We observed ΔE_p of about 300 mV for the untreated Au strip, and less than 100 mV after MESA treatment. Before starting ERGO fabrication process on the MESA treated Au substrate, we prepared 6.25 μg/ml of GO suspension from 0.1 mg/ml of stock solution. A mixture with a volume ratio of 2:1 of diluted GO and 1% Nafion (Step 4), respectively was prepared. About 5 μL of GO solution mixture was then dropcasted on the Au working electrode, followed by vacuum drying for 3 h (Step 5). Finally, electro-reduction of GO film was performed at -1.1 V with phosphate buffered saline (PBS) as the electrolyte for reduction. The electro-reduction was carried out for different time durations (1, 2, 3 and 6 h). For scaled-up preparation of electrode, we have developed a homemade set-up with PCB (printed circuit board) using multiple adapters that can produce multiple ERGO coated strips simultaneously, shown in Figure S1 (Step 6). Digital photographs of freshly prepared GO and ERGO coated Au strips are also shown in Figure S1(i). Scale bar corresponds to the actual dimension of electrodes (working, counter, and reference) which are patterned on the Au test strips. The geometrical surface area of the active working electrode (ERGO) was maintained as ~0.25 cm^2 for all the test strips. Morphology and chemical analysis of these electrodes were done by FESEM-EDX (Figure S1 (ii)) and XPS techniques (Figure 1 of the manuscript), respectively. We used these ERGO coated strips for further analytical measurements using cyclic voltammetry (CV), linear sweep voltammetry (LSV) and chronoamperometric (CA) techniques.

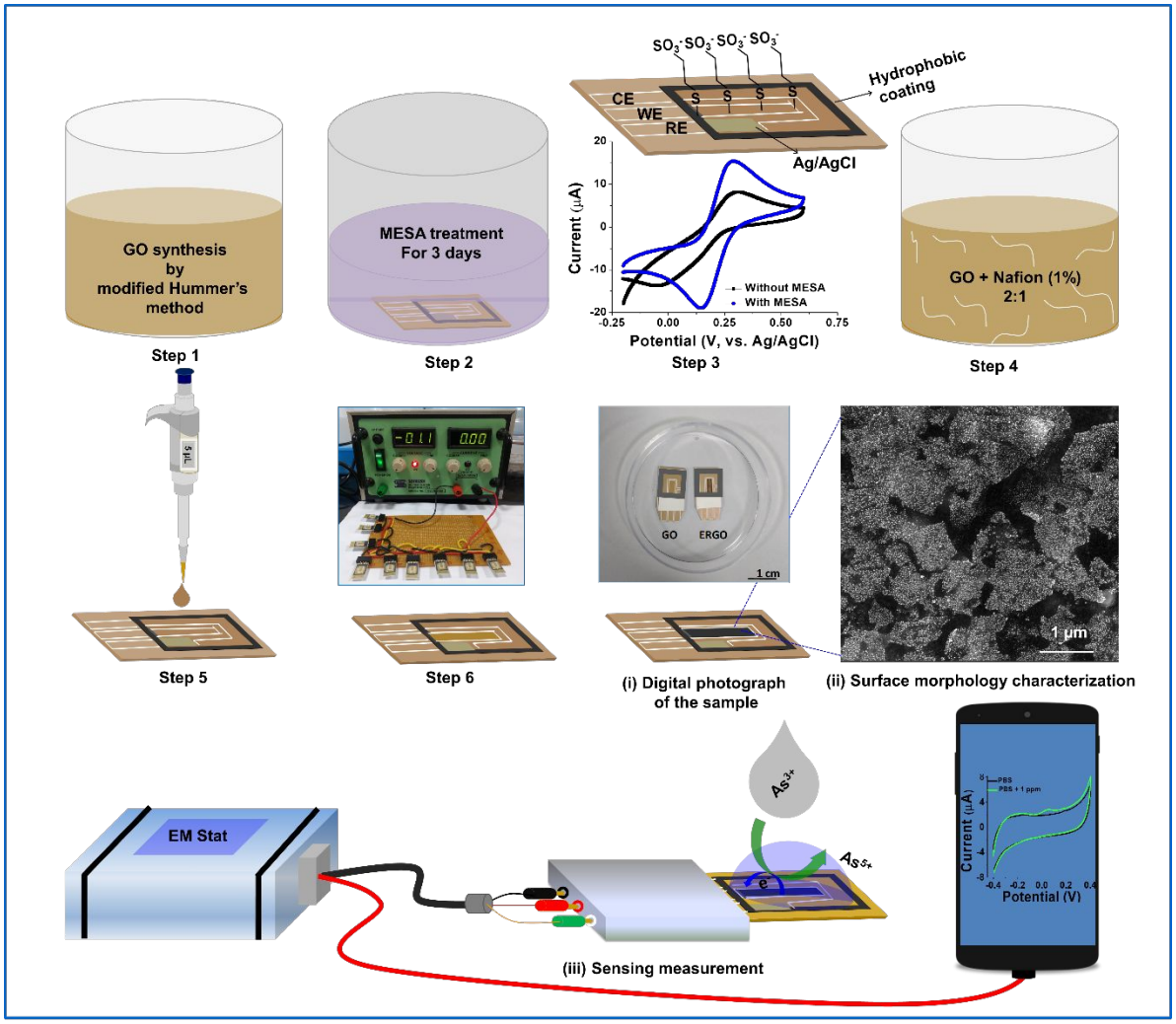


Figure S1. Schematic representation experimental steps involved in the fabrication of ERGO electrodes, steps involved in characterization using digital photography and microscopy, and finally, electro-analytical measurements for As^{3+} sensing. **Step1**: Wet chemical synthesis of graphene oxide (GO) by modified Hummer's method, **Step 2**: Pre-treatment of Au strips with sodium-2 mercaptoethane sulphonate (MESA) for decoration of self-assembled monolayer, **Step 3**: Au strip decorated with monolayer of MESA (top image) and cyclic voltammogram (bottom Figure) of Au strip before and after MESA treatment, **Step 4**: Dilution of as synthesized GO solution and mixture with Nafion (1% by weight) in 2:1 (by volume) ratio. **Step 5**: Dropcasting of solution mixture (5 μL) on working electrode (WE) of Au strip, **Step 6**: Electro-reduction of GO coated strip (bottom image) at -1.1 V using home-built setup (top

image), **(i)** Digital photograph of the electrode strips before and after electro-reduction (top image). The electro-reduced sample was used for further characterization (bottom image), **(ii)** Scanning electron microscopy (SEM) image of the as-prepared ERGO6 sample, and **(iii)** Sensitivity of ERGO6 electrode to As^{3+} was measured through cyclic voltammetry (CV) using EMSTAT (Plamsense) and corresponding voltammogram is shown in the right side image.

S2. SEM and EDS measurements of ERGO1 and ERGO6 samples

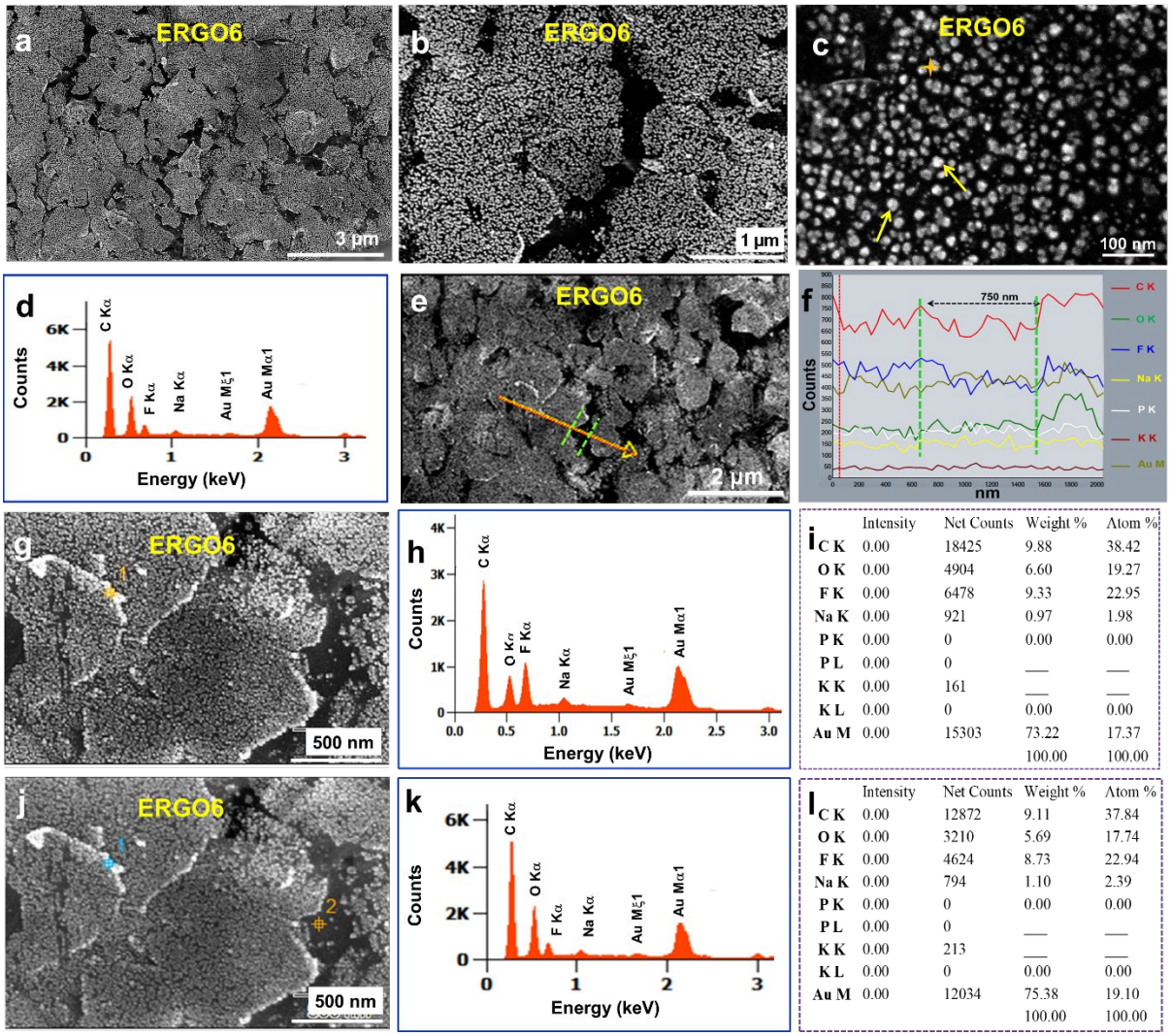


Figure S2. Morphological characterization and elemental analysis of as prepared ERGO electrodes. FESEM image of ERGO6 at different magnifications: (a) 3 µm, (b) 1 µm and (c) 100 nm. (d) EDS spectrum collected from a single nanosheets at the "+" marked point. (e) EDS line scan on the surface of ERGO6 in the direction of arrow. (f) Variation of possible elements of the electrode. Black portion on the electrode surface marked with dotted green lines indicates that there is a crack (width ~750 nm) on the electrode surface. In this region, the overall film thickness is less compared to other regions. In the deep region, the intensity of both carbon (C) and fluorine (F) is reduced, however, intensities of other elements is the same. (g) Point EDS spectrum was taken at the region marked as '1', (h) EDS spectrum at '1', (i) atomic percentage

of all the elements from (h). (j) Point EDS spectrum was taken at the region marked as ‘2’, (k) EDS spectrum at '2' and (l) atomic percentage of all the elements from (k).

FESEM images of ERGO6 at different magnifications are shown in Figure S2a-c. Figure S2d represents the EDS analysis of small nanosheets of ERGO6. The EDS spectrum was collected at the marked point (+) of Figure S2c. There are several microcracks present on the surface of ERGO6, and chemical composition of the film is uniform throughout the surface, as confirmed by elemental mapping through EDS line scan across the crack (Figure S2e-f). In the EDS line scan spectrum (Figure S2e), all major elements (C, O, and F) of ERGO6 were noted. However, a dip was observed in both carbon and fluorine (F) line profiles, which clearly indicated that cracks were formed within a few layers of ERGO6. Same EDX line profiles indicated that there might be a continuous ERGO film beneath the microcracks. Therefore, surface of ERGO6 was uneven as the edges of the smaller particles were exposed and the edges of the ERGO particles are marked with arrows as seen in an FESEM image (Figure S2c). Moreover, point EDX scan was performed on both nanoparticles-assembled sheet (marked on Figure S2g) and a microcracked portion of the sheet (marked on Figure S2j). The corresponding elemental analyses are shown in Figure S2h-i and Figure S2k-l, respectively. These results also confirm that both the electrodes are formed by stacking a few layers of thin ERGO sheets. However, the top layer of ERGO6 constitutes a large number of planar sheets, which are assembled from smaller nanosheets.

S3. XPS analysis of GO and ERGO electrodes

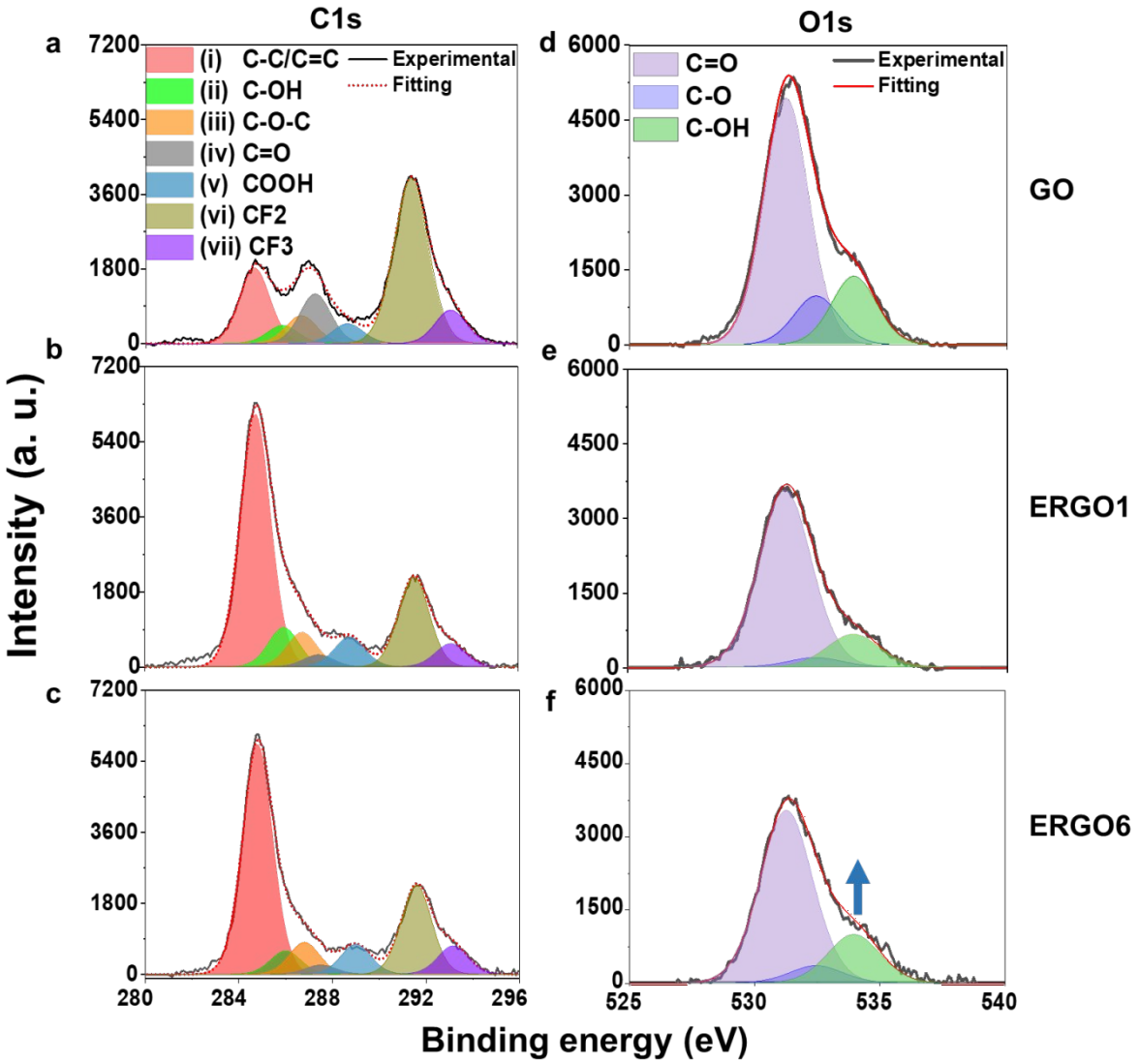


Figure S3. XPS C1s (a-c) and O1s spectra (d-f) for GO, ERGO1 and ERGO6, respectively. The blue arrow in (f) indicates an increase of C-OH intensity, which could be attributed to the oxidation at the edges of small ERGO nanosheets.

Table S1. Binding energies of both C1s and O1s for GO/ERGO electrodes

Binding energies of C1s (eV)	**Binding energies of O1s (eV)**
284.7 (C-C/C=C)	531.2 (O-C=O)
286 (C-OH)	532.4 (C=O)
286.8 (C-O-C)	533.9 (C-O)
287.5 (C=O)	
289.1 (COOH)	
292.6 (CF2)	
293.2 (CF3)	

The deconvoluted C1s are assigned as (i), (ii), (iii), (iv), (iv), (v), (vi), and (vii) in Figure S3a and their corresponding binding energies at 284.7, 286.0, 286.8, 287.5, 289.1, 292.6, and 293.2 eV are due to several functional groups on the basal plane of GO and ERGO samples. Peak (i) is responsible for carbon to carbon bonds in both sp^2 (C=C) and sp^3 (C-C) configurations, while (ii) and (iii) are assigned to C-O bond of hydroxyl (C-OH) and epoxide (C-O-C) functional groups, respectively. The peaks (iv) and (v) can be attributed to C=O bond of ester groups and carboxylic acid groups. The two peaks ((vi) and (vii)) of the spectra correspond to di-, and tri-fluorocarbon (CF2 and CF3) which are the main functional groups of Nafion binder used in electrode fabrication. Electrochemical reduction of GO minimizes the oxygen functional groups in the ERGO samples, resulting in the decrease of photo-emitted electron intensity associated with C-O bonds. Each O1s spectrum is decomposed into three peaks corresponding to O-C=O (~531.2 eV), C=O (532.4 eV), and C-O (533.9 eV) functional groups.

S4. Calculation of dielectric constant of ERGO1 and ERGO6 samples from impedance spectroscopy results

The dielectric constant of both ERGO1 and ERGO6 can be expressed as,[1]

$$\epsilon = \epsilon' - \epsilon''$$

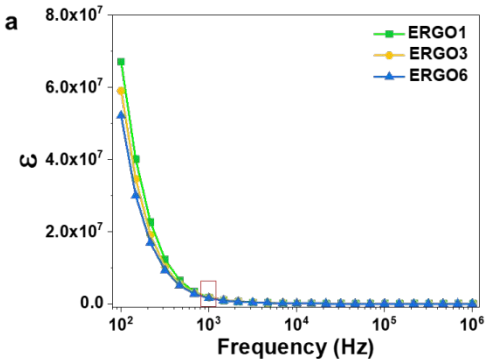
Where, $\epsilon' = \frac{Z''.l}{2\pi f \epsilon_0 A Z^2}$ and $\epsilon'' = \frac{Z'.l}{2\pi f \epsilon_0 A Z^2}$

The above equation can be simplified as, $\varepsilon = \frac{l}{2\pi\epsilon_0 A}\left[\frac{Z'' - Z'}{Z^2.f}\right]$

Here, Z' and Z'' are the real and imaginary parts of total impedance (Z) of all the samples obtained from Nyquist plot, f is the frequency maintained during EIS measurements, ϵ_0 is permittivity of the free space, A is the area and l is the thickness of the samples. The term, which is outside the bracket, is constant and same for all the electrodes.

S5. Some more results of impedance spectroscopy

a



b

Sample	Concentration of negative charges	Concentration of positive charges
ERGO1	7.8×10^{22} /cm^3	1.5×10^{20} /cm^3
ERGO3	9.2×10^{22} /cm^3	6.5×10^{22} /cm^3
ERGO6	0.1×10^{24} /cm^3	3.9×10^{27} /cm^3

c

Sample	R_s (Ω)				R_{ct} (Ω)			
	Before		After		Before		After	
	Exp.	Fit	Exp.	Fit	Exp.	Fit	Exp.	Fit
ERGO1	92	96	94	95	11	11	11	10.7
ERGO3	100	103	90	93	17	16	17	16.8
ERGO6	104	109	87	78	15	14	7	6.5

Figure S4. (a) Variation of dielectric constant of ERGO samples. Mott-Schottky measurements of all the samples were performed at a frequency of 1 kHz and the dielectric constant of all the electrodes at this frequency is the same as indicated in red shaded area. (b) Tabulated data of charge carrier concentration for all the ERGO electrodes. (c) Results obtained after fitting the impedance spectroscopy data and a tabulated data of interfacial resistances of each electrode before and after exposure to As^{3+}.

S6.

Table S2. Charge carrier density of different rGO samples including our ERGO electrode

Sample	Charge carrier	Carrier concentration	Measurement method	Reference
Thermally reduced graphene oxide (rGO) film	Electron	2.21 x 10^{20} /cm^3	Hall measurement	2
Laser annealed rGO film	Electron	4.7 x 10^{21} /cm^3	Hall measurement	3
Mechanically exfoliated graphene	Electron	7.5 x 10^{11} /cm^2	Back gated field effect method	4
Graphene formed by chemical vapour deposititon (CVD)	Electron	0.3-1.9 x 10^{12} /cm^2	Back-gated field-effect method	5
CVD graphene	Electron	0.4 x 10^{12} /cm^2	Electrolyte-gated field effect method	6
Electrochemica reduced graphene oxide (ERGO)	Positive and negative charge	~3.9 x 10^{27} /cm^3 (positive charge density) and ~10^{23} /cm^3 (negative charge density)	Electrochemical impedance spectroscopy: Mott-Schottky method	This work

S7. CV of ERGO samples with 1 ppm of As^{3+}

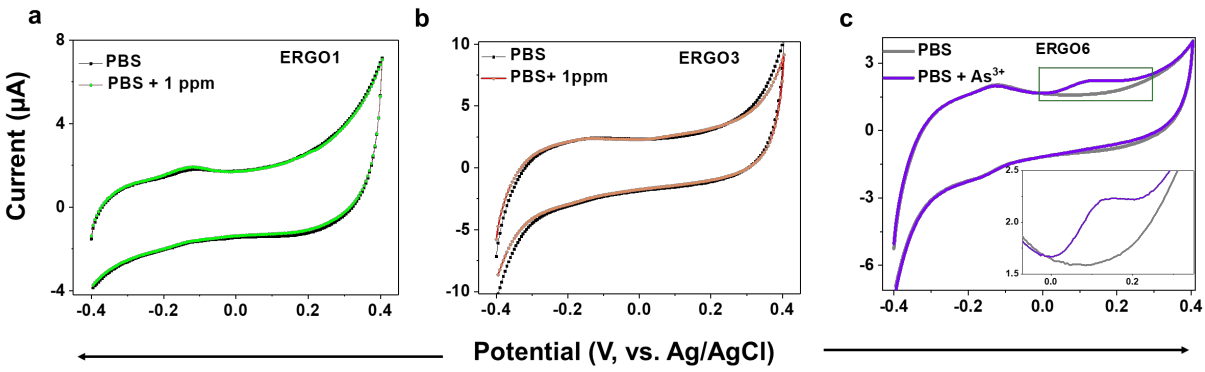
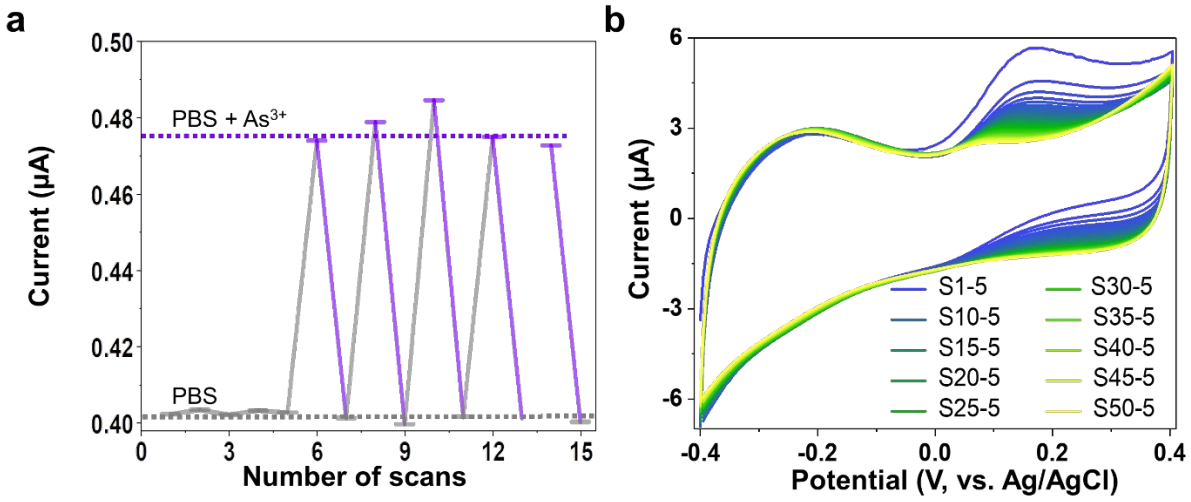


Figure S5. CV of (a) ERGO1, (b) ERGO3, and (c) ERGO6 using 1 ppm As^{3+} in PBS, inset figure shows the zoomed portion of the selected area of parent voltammogram.

S8. Ion chromatography measurements and experimental results



c

As3+ conversion			
Sample name	**Input sample**	**With / without CV**	**As5+ measured by IC (ppm)**
Blank		Without	0
1 ppm std		Without	1
3 ppm std		Without	3
5 ppm std		Without	5.01
Milli-Q		Without	0
Input	As3+ - 2.5 ppm	Without	0
Control	As3+ - 2.5 ppm	Without	0
10 cycles	As3+ - 2.5 ppm	With	0.61
20 cycles	As3+ - 2.5 ppm	With	0.88
40 cycles	As3+ - 2.5 ppm	With	1.49

Figure S6. (a) Chronoamperometric (CA) response of ERGO6 with 1 ppm As^{3+} in PBS (pH~7) with respect to the response current of PBS alone. CA was performed by keeping the potential fixed at +0.1 V and measurement was continued for 30 s in each scan. (b) Continuous CV scans for 50 cycles with 2.5 ppm of As^{3+} in DI water, and (c) summary of the ion chromatography (IC) measurements after electrochemical conversion of As^{3+}.

For IC measurement, we performed continuous CV scans for 50 cycles (voltammogram shown in Figure S6b) with 2.5 ppm of As^{3+} solution prepared in distilled water. Subsequently, we carried out IC measurement of the resulting solution and the chronogram is shown in Figure 3b in manuscript. In this Figure, we observed a peak at a retention time of ~23.25 s and this was assigned by measuring the chronogram with standard (STD) As^{5+} solutions of different concentrations. 1, 3 and 5 ppm of As^{5+} solutions were purged into the IC column and chronograms were recorded. We observed three peaks with different intensities at the same retention time. Therefore, the peak observed at the retention time of ~23.25 s corresponded to As^{5+} only. Furthermore, we investigated CV cycle number-dependent conversion of As^{3+} to As^{5+} and measured IC of the resulting solutions after a certain number of CV cycles. Here, we measured IC chronograms of the input solution (2.5 ppm of As^{3+}) after 10, 20 and 40 CV cycles and the results are shown in Figure S6b. The conductivities of three standard As^{5+} solutions (1, 3 and 5 ppm) were also measured along with the blank (solution without As^{5+}), control sample (2.5 ppm of As^{3+} solution) which was kept for overnight in air and three electrochemically oxidized solutions (after CV cycles). In the chronogram, it is seen that both blank and control sample do not show any peak at the retention time of As^{5+} (23.25 s), while three electrochemically oxidized samples showed three peaks of different conductivity at the same retention time. Figure S6c presents a summary of the IC data of the resulting solutions after electrochemical oxidation at ERGO6 electrode.

S9. Scan rate dependence of CV with As^{3+} and concentration dependence of CV using ERGO6 electrode

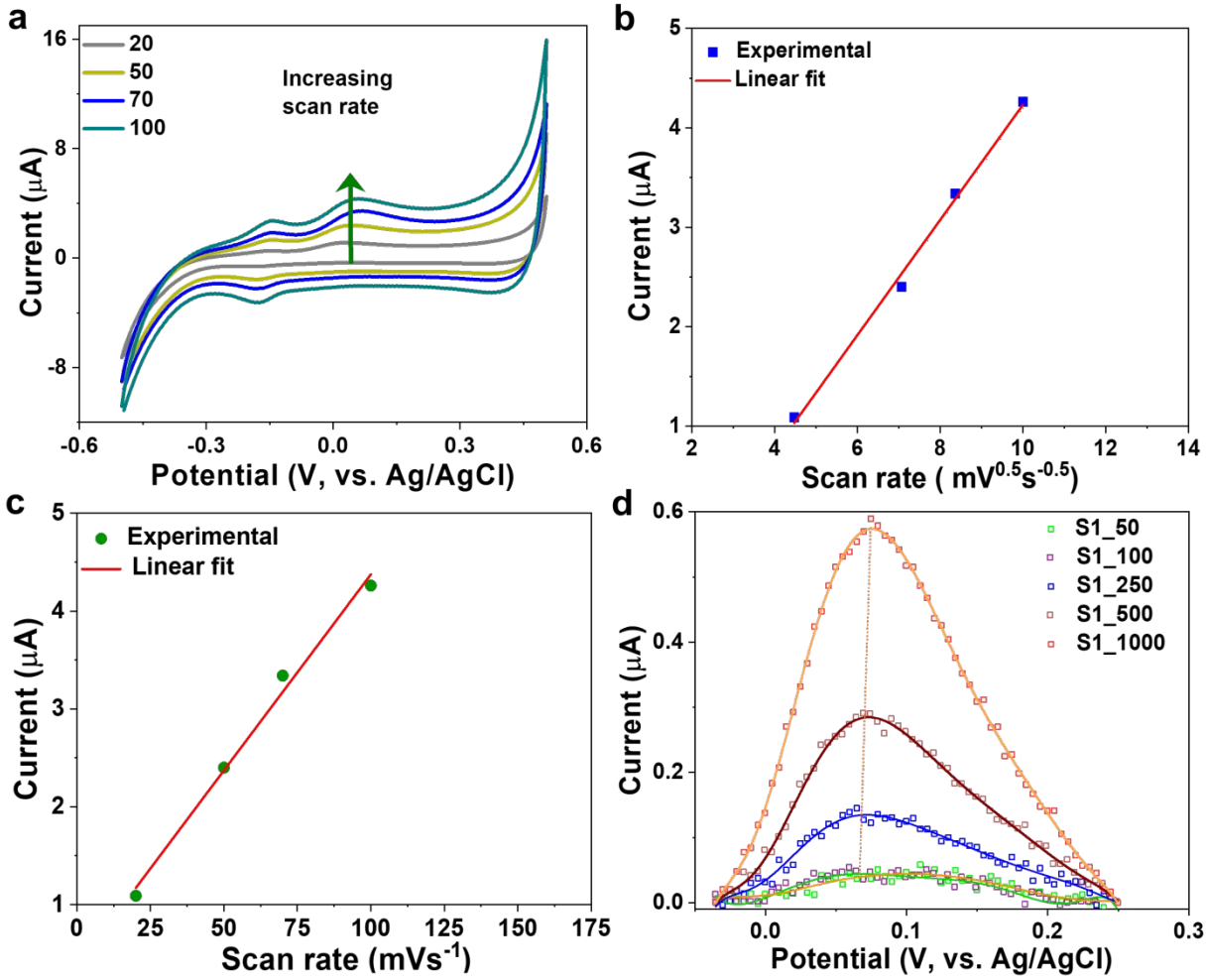


Figure S7. (a) Scan rate-dependent cyclic volammogram of ERGO6 using 200 ppb of As^{3+} in PBS. Variation of peak current with (b) square root of the scan rate, (c) scan rate with corresponding linear fits and (d) linear sweep voltammogram of ERGO6 with different concentrations (50 -1000 ppb) of As^{3+} at a fixed scan rate of 50 mVs^{-1}.

To understand the nature of As^{3+} interaction with the surface of ERGO6 and possible electron transfer at the ERGO6/electrolyte interface, we measured the CV at different scan rates (υ) with an aqueous solution of 200 ppb of As^{3+} (Figure S7a). We plotted the peak current i_p (A)

as a linear function of both $\upsilon^{1/2}$ (Randles-Sevcik equation) and υ, as shown in Figure S7b and S7c, respectively. Figure S7b represents the adsorption and subsequent oxidation of As^{3+} is a diffusion controlled process, while Figure S7c illustrates the surface adsorbed species (As^{3+}) on ERGO6.

Linear sweep voltammetry (LSV) measurements were performed with different concentrations of As^{3+}, the peak at ~ +0.1V in the voltammogram is the current response corresponding to As^{3+} oxidation. The voltammogram shown in Figure S7d corresponds to the increasing As^{3+} oxidation with gradual increase of As^{3+} concentration and the response is linear.

S10. Concentration dependent chronoamperometry measurements in PBS and field water

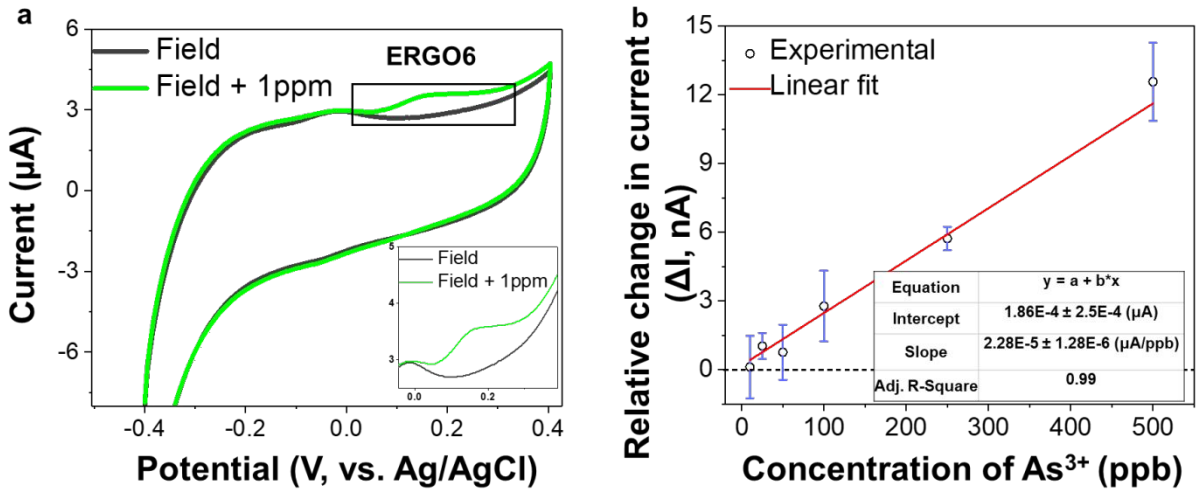


Figure S8. (a) CV performed with the same ERGO6 electrode with field sample (F1), inset figure shows the zoomed curve of the selected area of the parent voltammogram. (b) Measurement of the calibration curve to determine the LOD in field sample.

S11. Interference measurements of bare ERGO6 and bare Au electrode with different cations and anions

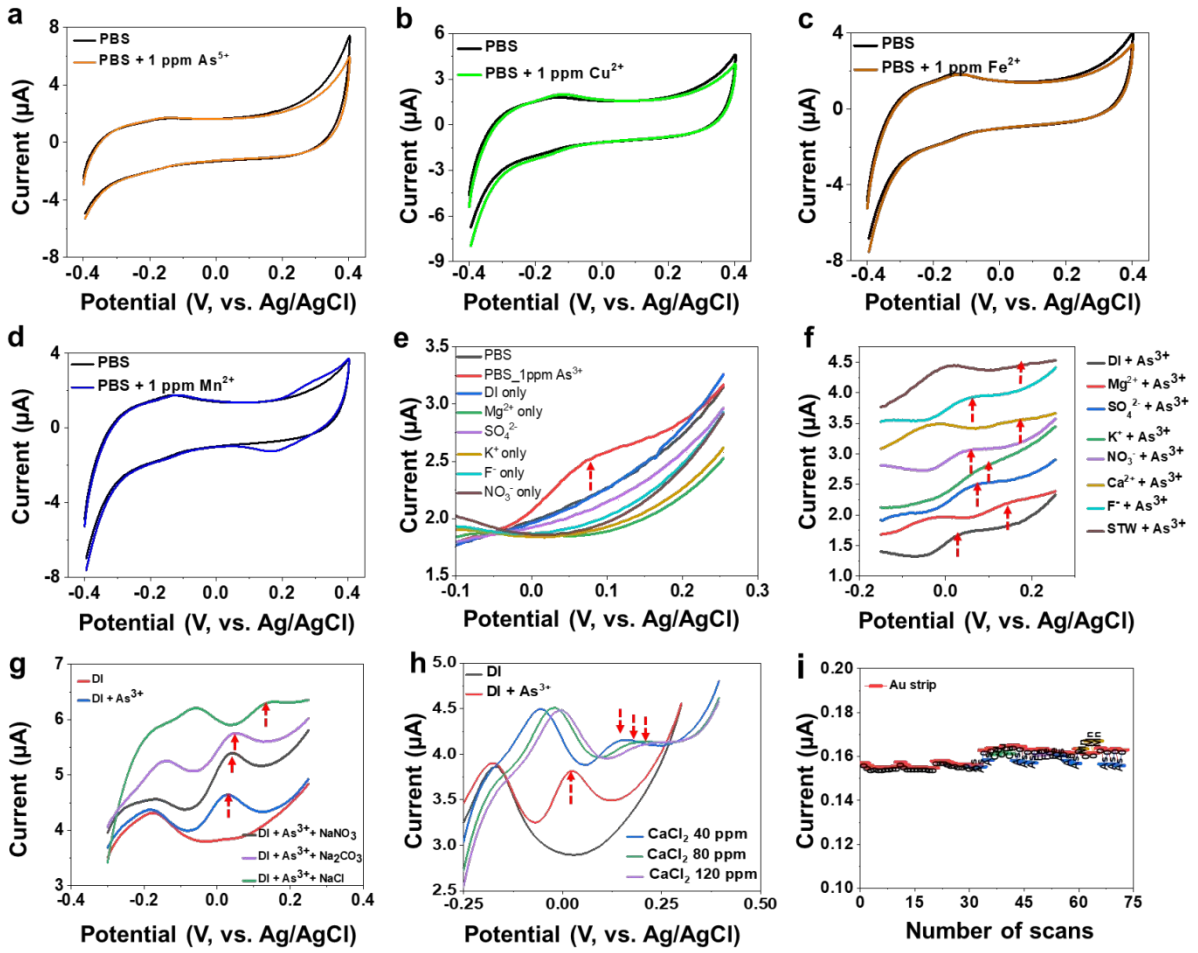


Figure S9. CVs of ERGO6 in presence of different heavy metal ions (a) As^{5+}, (b) Cu^{2+}, (c) Fe^{2+}, and (d) Mn^{2+}, respectively. CV of ERGO6 to study the (e) responsivity of several cations and anions separately without As^{3+}, (f) effect of some other cations and anions on As^{3+} response, (g) effect of different anions on the As^{3+} response (in presence of Na^{+}), (h) effect of Cl^{-} concentration on the As^{3+} response (in presence of Ca^{2+}). Concentration of the cations was maintained similar to their concentration in simulated synthetic tap water. Concentration of As^{3+} was maintained at 1 ppm in all the cases. Red dotted arrow line indicates the response of arsenic. For clarity, we have shown oxidation cycle of the each CV. (i) Chronoamperometry current of pristine Au strip in presence of Cu^{2+}, Fe^{2+}, and Mn^{2+} along with As^{3+}. Concentration of each ion was maintained at 1 ppm.

Figure S9a-d represent the CVs of ERGO6 in presence of four major heavy metal ions and the experiment was carried out in PBS. In the voltammograms, there was no peak observed in the potential window (-0.4 to +0.4 V) due to any of the ions. Oxidation (+0.3 V) and reduction peaks (+0.17 V) of Mn^{2+}, did not interfere with the chronomaperometric response of As^{3+}, as the peak positions were different compared to As^{3+} (~+0.1V)

We have further studied the electrochemical response (CV) with a few more cations (Ca^{2+}, K^{+}, and Mg^{2+}) and anions (F^{-}, SO_4^{2-}, CO^{3-}, Cl^{-} and NO_3^{-}) of relevance in order to prove the selective response. The results are shown in Figure S9e. ERGO6 responded to As^{3+} only and there was no response towards other ions. We used three common anions (F^{-}, SO_4^{2-}, and NO_3^{-}) and a common counter cation (Na^{+}), while, for Cl^{-} anion, two counter cations (Ca^{2+} and Mg^{2+}) were used. The concentrations of both cations and anions were chosen as per standard concentrations used for the preparation of synthetic tap water (STW) as mentioned in the supporting information of S. Mukherjee et al.[7] Concentrations of cations and anions used during the experiments are shown in Table S3

. The results are shown in Figure S9f and As^{3+} response was seen in all cases. The peak corresponding to As^{3+} oxidation shifted to a higher potential in presence of Ca^{2+} and Mg^{2+}. Also peak height decreased for the two ions. The presence of high concentrations (40 to 120 ppm) of Cl^{-} passivated the ERGO surface and formed an extra electrical double layer, so that charge transfer kinetics due As^{3+} oxidation was slower. This could lead to shifting of oxidation peaks to higher potential and diminishing of the corresponding peak height. To verify the hypothesis, we performed CV with different anions (NO_3^{-}, CO_3^{-}, and Cl^{-}) in presence of 1 ppm of As^{3+} and the same counter cation (Na^{+}) was chosen. The results are shown in Figure S9g in which As^{3+} response was detected for all the anions. In presence of Cl^{-}, As^{3+} reduction peak shifted to higher potential with diminished peak height. For further confirmation, CV was performed with As^{3+} in presence of different Cl^{-} concentrations. On increasing of Cl^{-}

concentration, the peak gradually shifted to higher potentials and the peak height diminished gradually with increasing Cl^- concentrations. The results are presented in Figure S9h. To verify that ERGO6 is exclusively responsible for As^{3+} sensing, we carried out a control experiment by measuring the CA current of bare Au strip with four metal ions (As^{5+}, Cu^{2+}, Fe^{2+}, and Mn^{2+}) including As^{3+}. The result is shown in Figure S9i. The concentration of each ion was maintained at 1 ppm. It was seen that Au did not show any response toward these metal ions.

S12. Concentrations of cations and anions in synthetic tap water

Table S3. Concentration of cations and anions used in the interference study of ERGO6 electrode

Cations / Anions	Concentration (ppm)
Na	63.6
Cl	87
Mg	14.34
SO_4	32.41
F	0.57
Ca	28.72
CO_3	43.22
NO_3	1.84

S13. Chronoamperometry measurements with various field samples

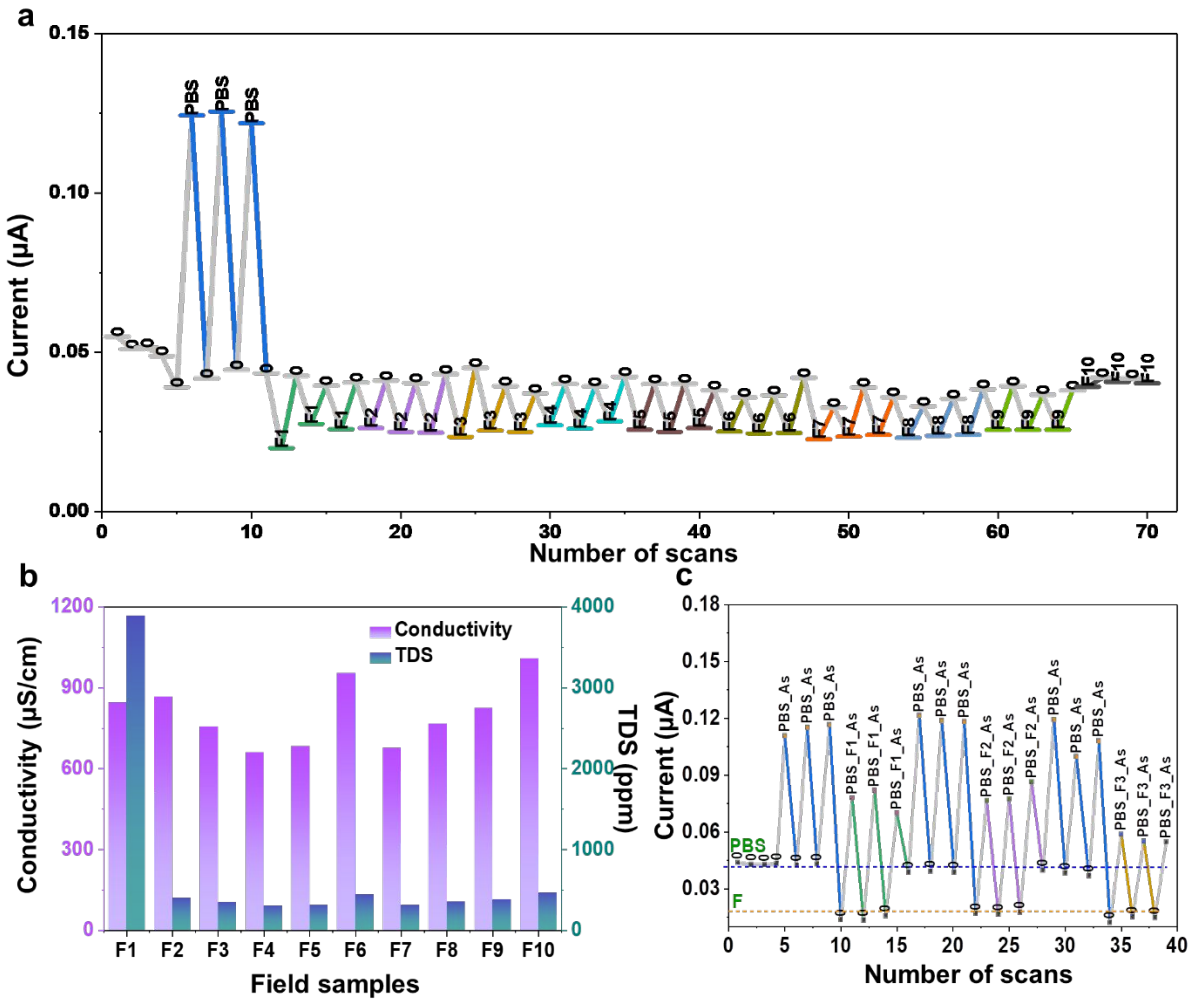


Figure S10. (a) Repetitive CA current response of ERGO6 with different field samples (marked as F1-F10) of different conductivity and TDS levels. CA response of various field samples (no arsenic was found). We tested with ten individual field samples. Each field sample was measured thrice. The first three samples in PBS had 1 ppm As^{3+}. (b) Variation of conductivity and TDS of all field water samples. (c) CA current response of 1 ppm As^{3+} spiked PBS and different field samples. Initial three cycles represents current response of As^{3+} in PBS, while each three sets of the rest of the cycles represent current response of the electrode with a particular field sample having no As^{3+}. This experiment was carried out with three different field samples (F1-F3). Here, F stands for field water sample using same strip.

Figure 10a presents CA responses of different field water samples. All the field water samples were collected from different locations of South 24 Parganas district of West Bengal, India. Before starting the CA measurements with field water samples, total As concentration of all the water samples was measured by ICPMS and no As (less than 1 ppb) was found in these water samples. The current responses of all the field water samples were measured with respect to the current response of blank PBS. Before starting the CA measurements with field water samples, As^{3+} response was tested with ERGO6 strip for three scans followed by the same measurements performed with different field samples. It was seen that ERGO6 showed lower current response in presence of field sample with respect to the current response of blank PBS. The amplitude of the current response of all the field samples was similar, although there were differences in conductivity and TDS. Variation of TDS and conductivity of all the field samples is shown in Figure S10b. Negative response by ERGO6 strips with field water samples was owing to the presence of Cl^- in the field samples, as we have seen already in Figure S9h. The concentration of Cl^- ions in the field samples was around 1- 2 ppm. Cl^- ions might passivate some of the active sites of ERGO6, thus decreasing overall ionic current of the electrode in field samples compared to PBS. In addition, arsenic response of ERGO6 strip was checked in both PBS and field samples with spiking of 1 ppm As^{3+}. The results are depicted in Figure S10c. As^{3+} response was observed in both cases; however, the amplitude of the current response of As^{3+} in field water sample (~ 60 nA) is lower than the response in PBS (~ 70 nA).

S14: Raman spectra of ERGO6 measured at different scan depths in presence of water

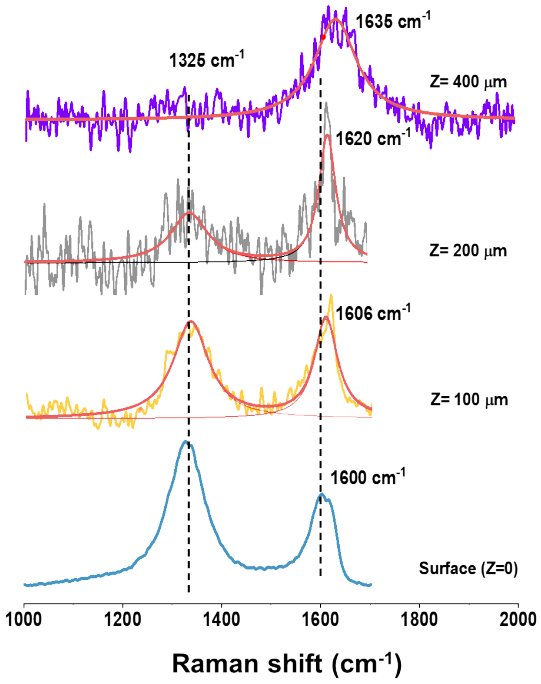


Figure S11: Raman spectra of the same electrode at different scan depths (Z) with respect to the surface of the electrode and measurement was carried out in presence of water.

S15. Gaussian fitting of G band of Raman spectra of ERGO6 without and with DI water

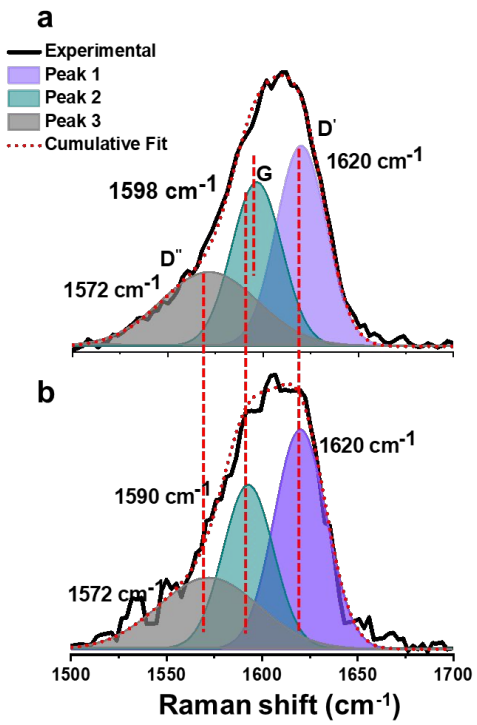


Figure S12. Fitting of G band of Raman spectra of ERGO6 electrode (a) with, and (b) without DI water.

S16. In-situ SPEC measurements of ERGO1 and ERGO6 in presence of only DI water, without and with external potential

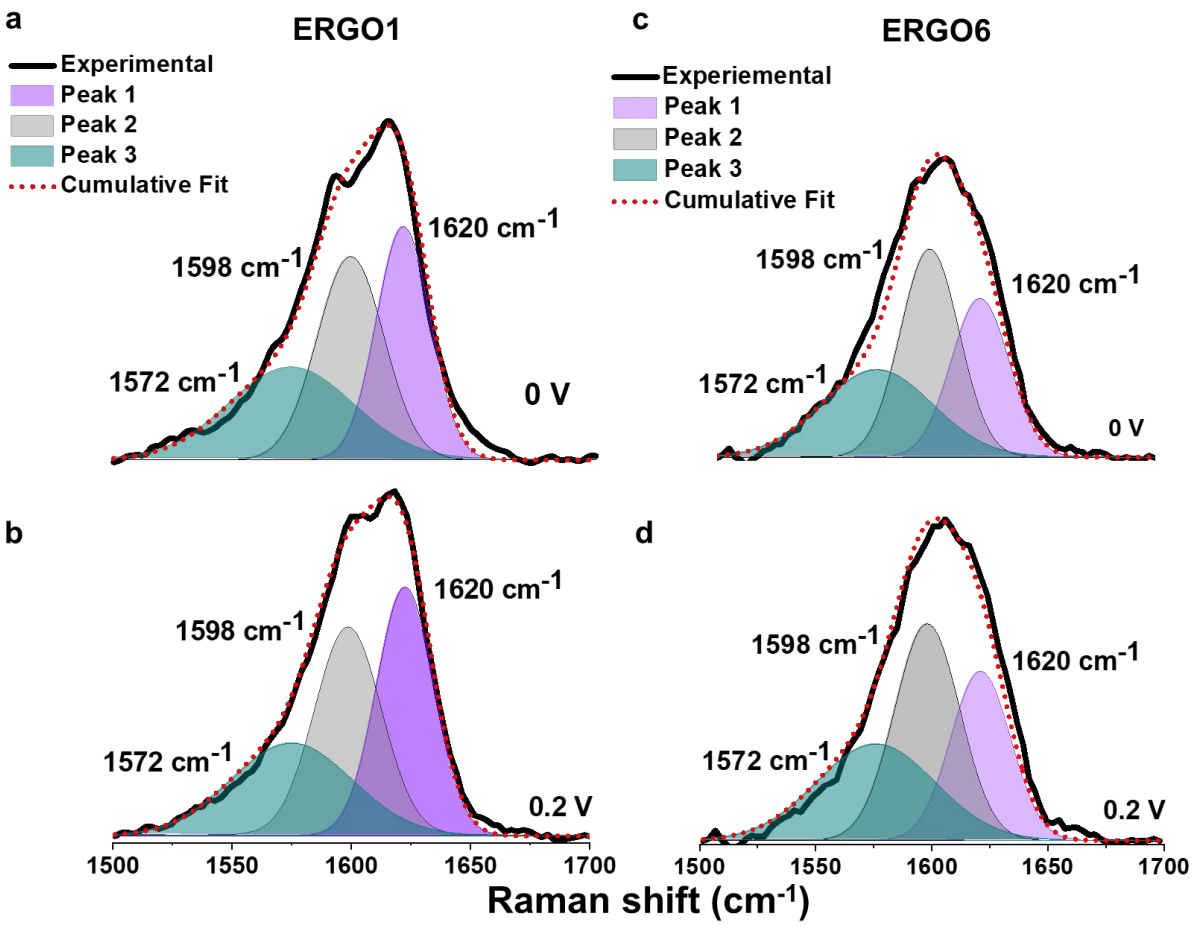


Figure S13. Gaussian fitting of G band of Raman spectra of ERGO1 electrode in presence of DI water acquired at (a) 0 V, and (b) 0.2 V. Gaussian fitting of G band of Raman spectra of ERGO6 electrode in presence of DI water acquired at (c) 0 V, and (d) 0.2 V.

S17. Chronoamperometry profiles of ERGO1 and ERGO6 without and with As^{3+} during in-situ spectroscopy measurements

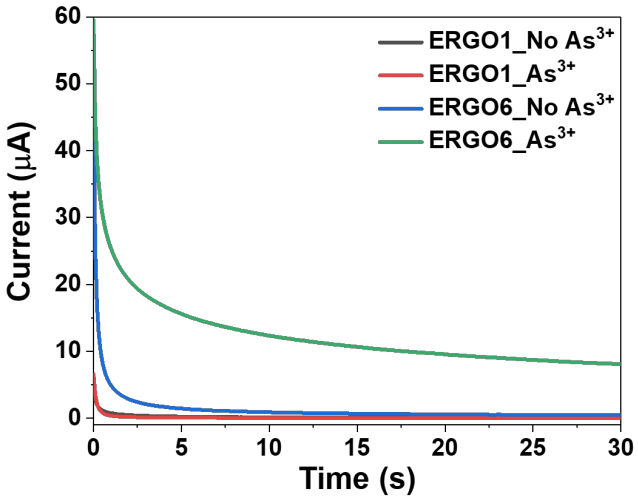


Figure S14. CA current of both ERGO1 and ERGO6 at 0.2 V in absence and presence of 1000 ppm of As^{3+}.

S18. Study of electrochemical stability of the electrodes

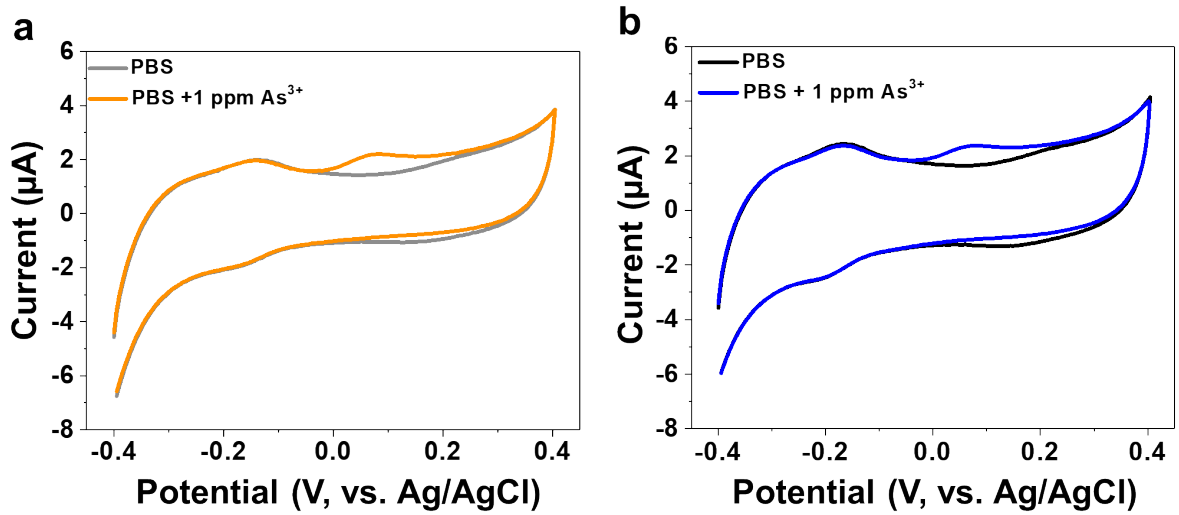


Figure S15. CV with 1 ppm As^{3+} using (a) as prepared ERGO6 and (b) the same strip after 2 months.

S19. Effect of potential on the G band of in-situ Raman spectra

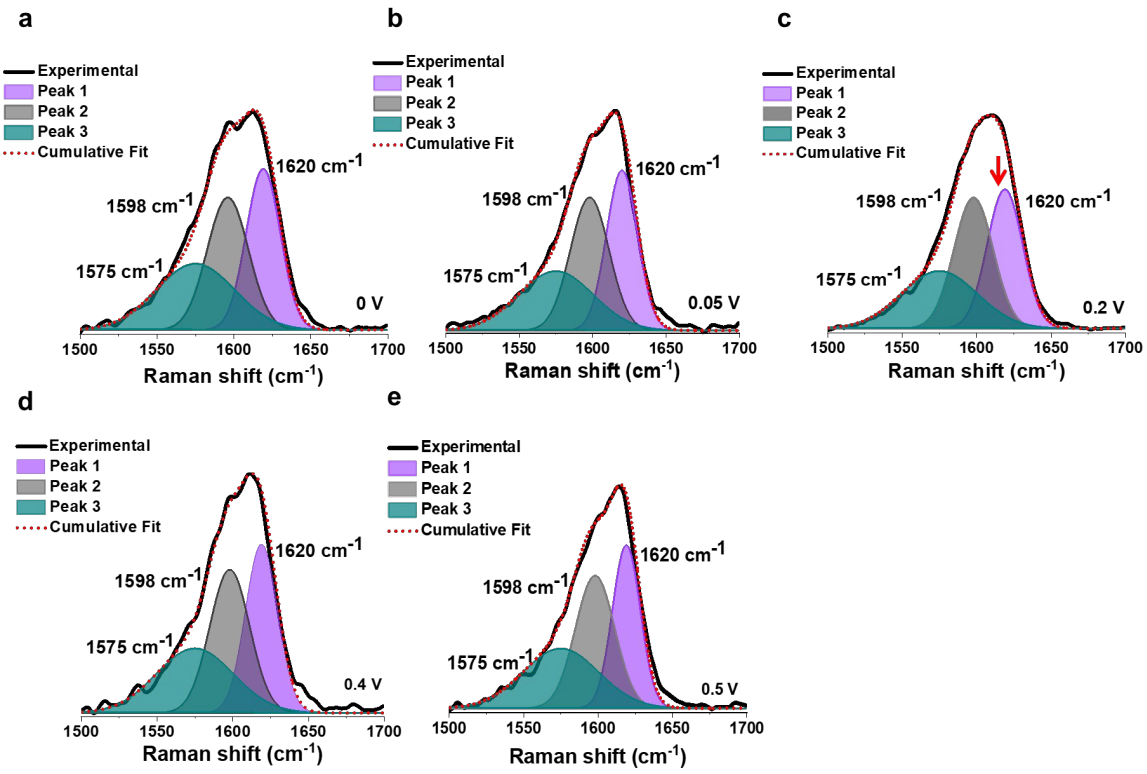


Figure S16. Gaussian fitting of G band of Raman spectra of ERGO6 in presence of As^{3+} acquired at different potentials: (a) 0 V, (b) 0.05 V, (c) 0.2 V, (d) 0.4 V, and (e) 0.5 V.

S20. In-situ spectroelectrochemical study of ERGO6 in presence of different cations including As^{3+}

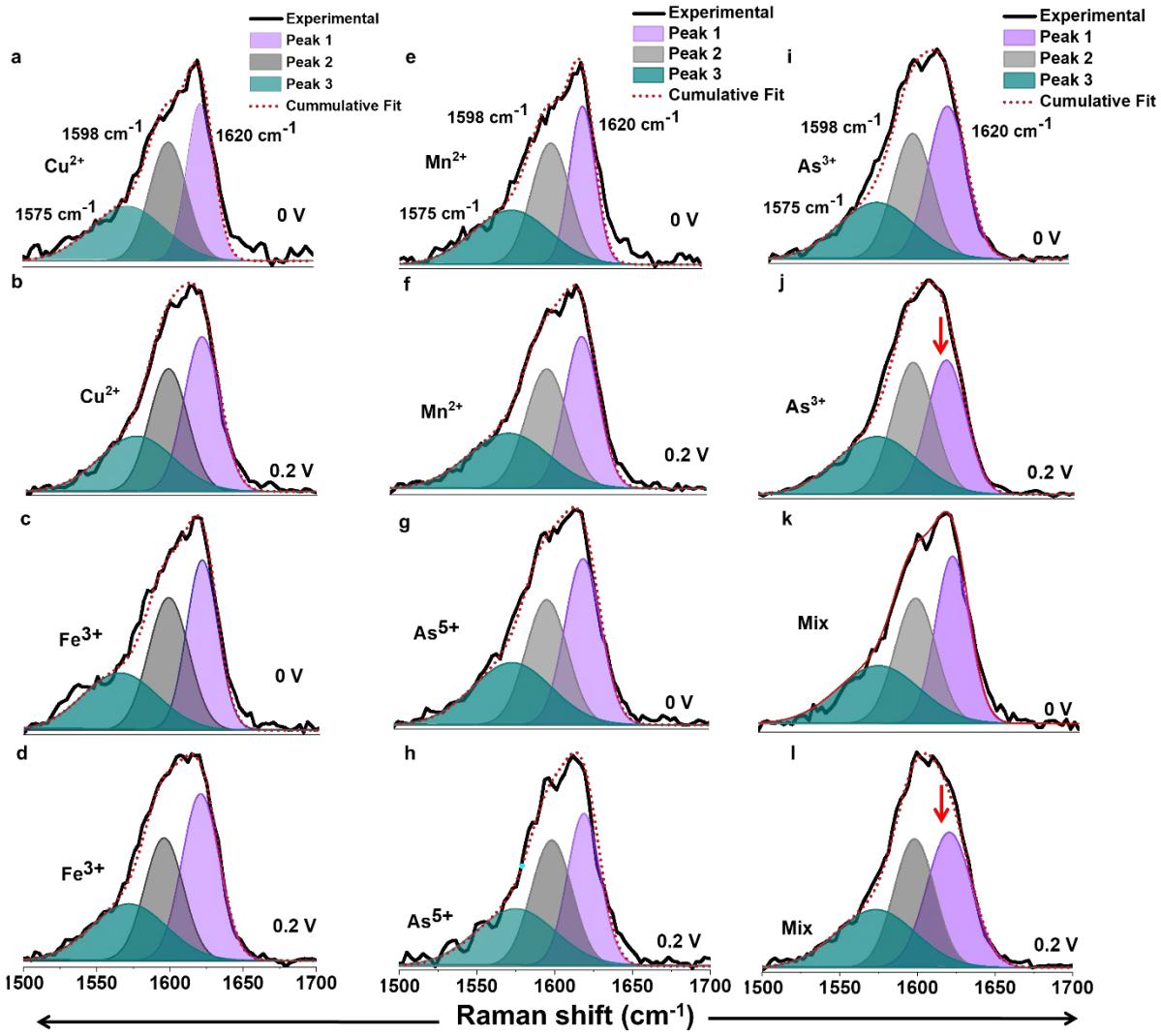


Figure S17. In-situ Raman spectra (G band) of ERGO6 electrodes in presence (a-b) Cu^{2+}, (c-d) Fe^{3+}, (e-f) Mn^{2+}, (g-h) As^{5+}, (i-j) As^{3+}, and (k-l) mixture of ions. Spectra were recorded for each ion at 0 V and 0.2 V. Concentration of each ionic species was maintained at 1000 ppm.

S21. FTIR spectra of ERGO6 and ERGO1 electrodes with and without As^{3+} and after electrochemical oxidation

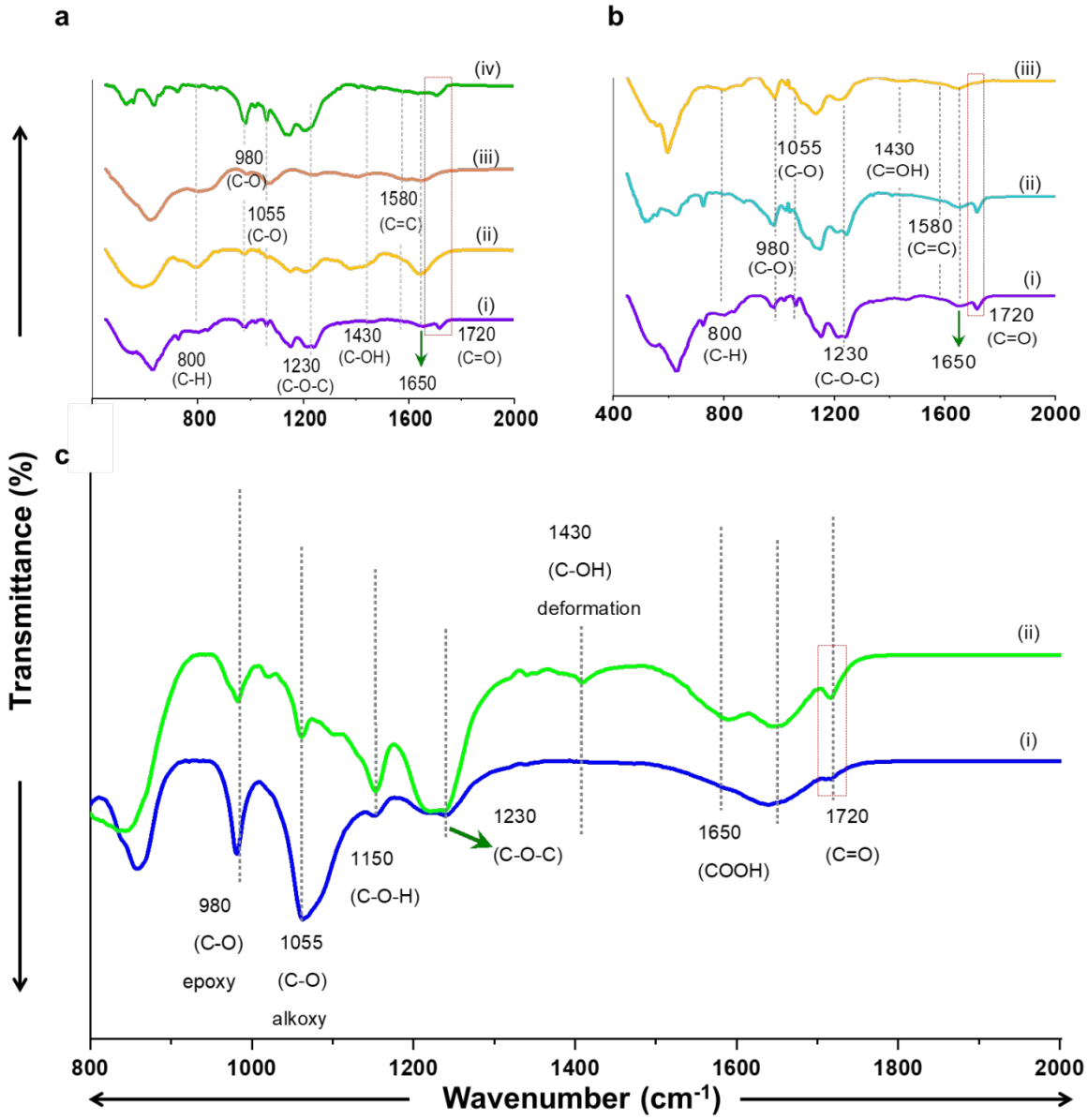


Figure S18. (a) FTIR spectra of the same strip without As^{3+} (purple trace), with As^{3+} (yellow trace), after washing with DI water (orange trace), and after electrochemical oxidation (green trace). (b) FTIR spectra of bare ERGO6 electrode (purple trace), same electrode with mixture of interfering ions (blue trace) and interfering ions along with As^{3+} (yellow trace).

(c) FTIR spectra of ERGO1 with (1000 ppm) and without As^{3+}.

S22. Computational Methodology

We used the Avogadro program package [8] for building up the structures of the analyte ions and the ERGO-analyte adducts, and for structure visualization and analysis, and for our UFF force field simulations. The Visual Molecular Dynamics (VMD) package was used to visualize and render the images of the structures.[9]

The geometry optimization of the $As(OH)_3$-ERGO adduct isomers 1 to 6 shown below were performed using all-electron density-functional theory DFT at the B3LYP/6-31G* level of theory in the neutral-charge state using the NWChem 7.0 package.[10] Among the six isomers, we have shown lowest energy isomer (isomer 2) in the manuscript (Figure 5a). For reasons of computational efficiency for all our other calculations, we used the grid-based projector augmented wave method as implemented in the GPAW software package.[11] We employed the linear combination of atomic orbitals (LCAO) mode of GPAW with a double-zeta plus polarization (DZP) basis set with GGA-PBE exchange-correlation functional[12] for all calculations. The valence electronic configuration of the GPAW setups were the default valence electronic configurations of those elements. A grid spacing of 0.2 Å was used, and the simulation box size was 32 Å for all calculations regardless of the model size for the purpose of being able to comparing Kohn-Sham eigenvalues between different molecular systems. During geometry optimizations, the stopping criterion for the maximum force on atoms was 0.04 eV/Å. To accelerate electronic structure convergence of ERGO and analyte molecules, we used a Fermi broadening of 0.02 eV for the electronic occupations. For the calculations involving Mn and Fe atoms in $3d^5$ configuration, we used spin-polarized DFT and set the magnetic moment to be constrained to its initial value of $\mu=\sqrt{n(n+2)} = 5.91\mu_B$, where the number of unpaired electrons, n, is 5, which corresponds to total spin S=5/2, and spin multiplicity of 6.

In our simulations, we neglect all solvent effects, both implicit and explicit, except where we include aqua ligands as part of the solvated species of the Fe^{2+}, Cu^{2+} and Mn^{2+} ions. Furthermore, our simulations are performed neglecting temperature, potential and electric field, and dynamical effects in our simulations. The positive surface-charge density of ERGO6 electrode at constant potential has been simulated approximately, by charging the model in DFT. For structural property, we used neutral model of ERGO as it is sufficient for bonding properties.

We describe our detailed simulation methodology in three stages, (1) Site-selectivity and binding of arsenite and other analytes to ERGO, (2) Oxidation mechanism of arsenite with ERGO (3) Ion-selectivity mechanism, in sections S22, S23, and S24, respectively, below. Additional results are also presented in the relevant section.

S23. Methodology of generation of ERGO models and binding of arsenite and analyte ions to ERGO

We now describe the process of (a) generating structural models of ERGO, (b) optimizing As-$(OH)_3$-ERGO adduct isomers and (c) Fe, Cu, Mn, and $H_2PO_4^-$ adduct isomers and binding energy study, in the respective sections below.

(a) ERGO models 1, 2 and 3

We first generated a rectangular-sheet model of ERGO (model 1) containing a total of 85 atoms of dimensions 7x10 Å ($C_{46}O_{19}H_{22}$), with a realistic oxygen/carbon surface atom ratio[13] using the MakeGraphitics[14] package which generates reduced graphene oxide models of the Lerf-Klinowski[15] type. The model contained four different types of functional groups – epoxide, hydroxyl (OH) groups, and C-O-C lactone groups and carboxylate (COOH) groups. The dimensions of the model 1 were 7×10 Å. The size of the model was chosen to be as small a possible but with realistic number of functional groups to model the analyte ion-ERGO interactions. Model 1 was then geometry-optimized using all-electron density-functional theory DFT at the B3LYP/6-31G* level of theory in the neutral-charge state using the NWChem 7.0 package[10]. The optimized geometry of model 1 is shown in Figure S19, below.

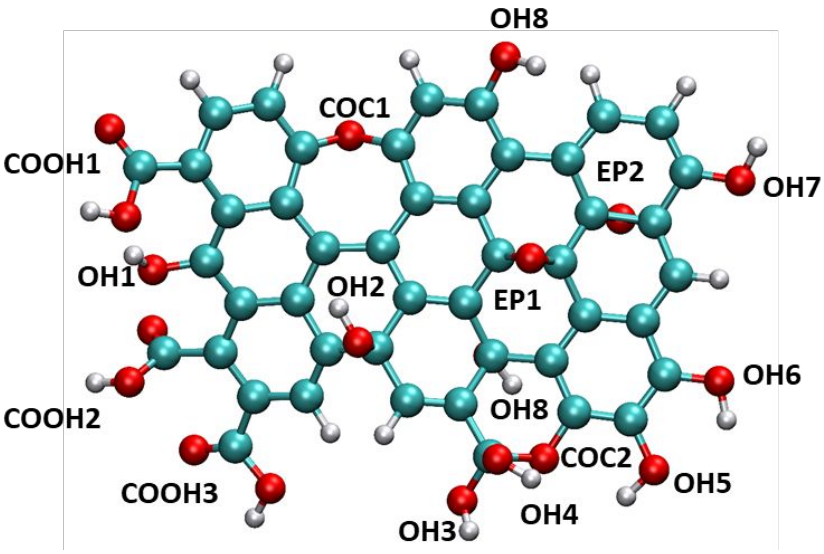


Figure S19. Model 1 – DFT-optimized structure of ERGO rectangular sheet with 85 atoms ($C_{46}H_{22}O_{17}$). The model has two pairs of epoxide and OH groups on the surface, and three COOH groups, and eight hydroxyl groups. The functional groups carboxylate (COOH), hydroxyl (OH), COC(lactone) and expoxide (EP) are labelled as indicated in brackets. Atom sphere colors are cyan for carbon, red for oxygen and white for hydrogen.

We created additional models of ERGO which were used in the ion-selectivity study, models 2 and 3, and their structures are shown in Figures S20 and S21, below. Model 2 was an ERGO sheet model with a lower ratio of edge functional groups and model 3 is a hexagonal flake model, and these were also optimized in the neutral and +1 charge states.

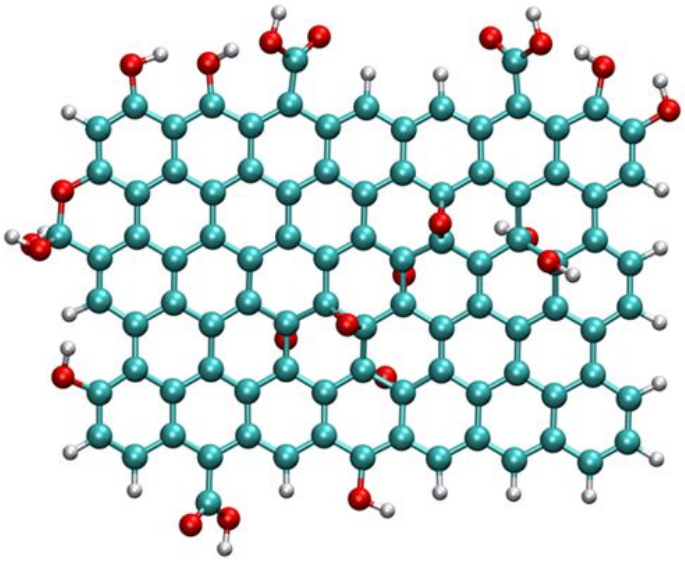


Figure S20. Model 2- DFT-optimized structure of a larger rectangular sheet with 144 atoms, ($C_{91}O_{22}H_{31}$). The model has three epoxide/OH group pairs on the surface pair, and the size of the model is ~12x18 Å. Atom sphere colors are cyan for carbon, red for oxygen and white for hydrogen.

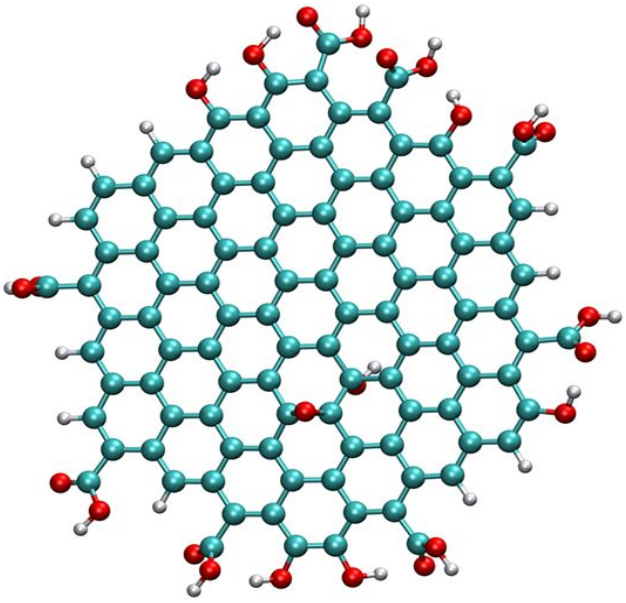


Figure S21. Model 3 - DFT-optimized structure of a hexagonal flake model of ERGO ($C_{101}O_{22}H_{30}$) with 153 atoms. The model has a single surface epoxide/OH group pair, and edges are decorated with 8 OH and 8 COOH groups. The size of the model is ~10 Å. Atom sphere colors are cyan for carbon, red for oxygen and white for hydrogen.

(b) $As(OH))_3$-ERGO adduct isomers 1 to 6

We investigated the binding-site selectivity of the arsenite molecule $As(OH)_3$ to the various functional groups on the surface and edges of ERGO sheet model 1. Geometry optimizations were carried out using NWCHEM with the $As(OH)_3$ molecule initially placed near to different functional group environments on the surface and edge of model 1, and in different orientations, with the As facing towards and away from the functional group. We obtained a set of six lowest-energy isomers, named as isomers 1 to 6, the structure of isomer 2 is shown in Figure 5 and structures of remaining isomers are shown in Figure S22. The types and number of bonds, and their lengths, are shown in Table S4 below. The additional covalently bonded adduct isomer structure with an As-O bond shown in Figure S22 was optimized using GPAW using the same calculation methodology described below.

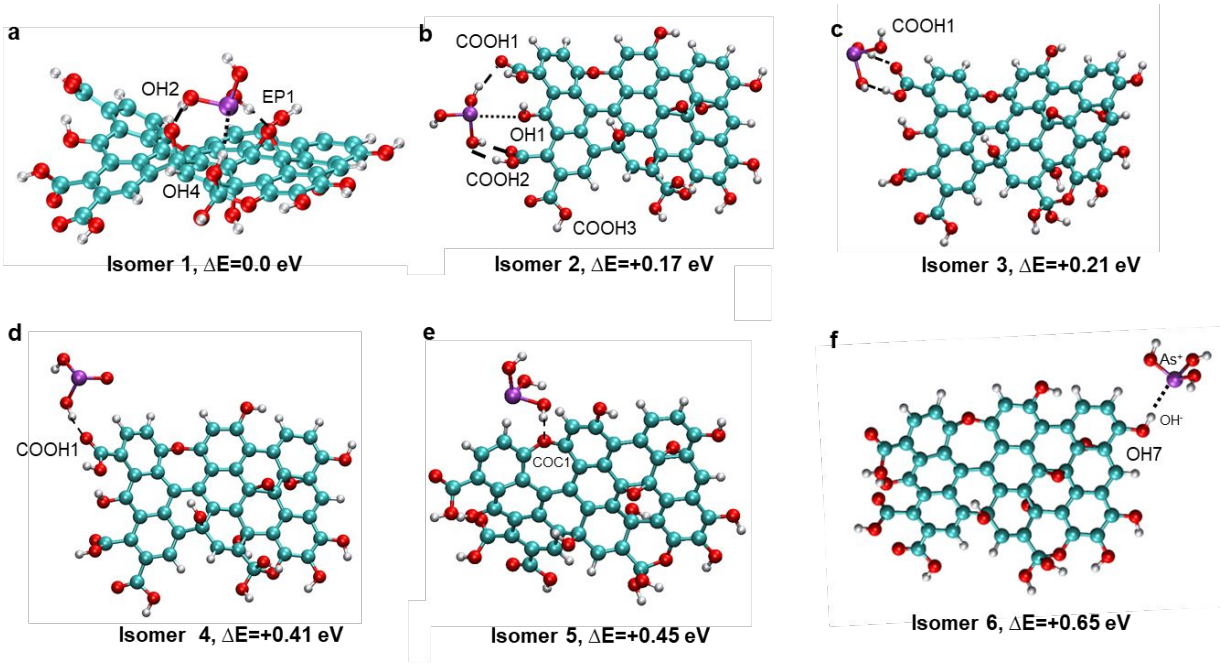


Figure S22. $As(OH)_3$ binding at different sites of ERGO model from DFT calculations. Isomers 1 to 6, showing binding possibilities of the As^{3+} species to the functional groups present on the surface and edges of the ERGO model. (a) Side view of the lowest-energy isomer 1 showing $As(OH)_3$ on the GO surface bound by two hydrogen bonds (HBs), indicated by dashed lines, between $As(OH)_3$ and ERGO epoxide (EP1) and hydroxyl group (OH2), and also a weak As^{+}-O^{-} interaction, indicated by dotted lines, with a surface hydroxyl group (OH4). (b) Isomer 3, binding to the COOH1 group by two HB's. (c) Isomer 4, binding to the COOH1 group via a single HB. (d) Isomer 5 involves binding via a single hydrogen bond to an edge C-O-C group. (e) Isomer 6, which is the highest-energy isomer has a weak As^{+}-O^{-} interaction with a hydroxyl group (OH7) and no HBs. The functional groups interacting with the $As(OH)_3$ are labelled according to Figure S23. The color scheme for the atomic spheres is purple for arsenic, cyan for carbon, red for oxygen, and white for hydrogen.

Table S4. Binding sites of $As(OH)_3$ on ERGO, the number of hydrogen bonds (HBs) and As^+-$(OH)^-$ interactions, bonding distances, and relative energies of isomers 1 to 6. The functional groups are labelled as in Figure 7 in the manuscript.

Isomer	Binding Sites on ERGO	No of HBs/As^+-$(OH)^-$ interactions	Bonding Distances/Å	Total Energy/a.u.	Relative Energy/eV
1	Surface OH2 and OH4 groups and epoxide group (EP1)	2/1	As---OH4: 2.55 HB(OH2): 1.91 HB(EP1): 1.88	-5657.099479	0.0
2	Edge COOH1 and COOH2 groups and OH1 group	3/1	HB(COOH1): 1.93 HB(COOH1): 1.89 HB(COOH2): 2.16 As---OH1: 3.26	-5657.093368	0.17
3	Edge COOH1 group	2/0	HB(COOH1): 1.71 HB(COOH1): 1.82	-5657.091603	0.21
4	Edge COOH1 group	1/0	HB(COOH1): 1.88	-5657.08450	0.41
5	Surface C-O-C group	1/0	HB(COC1): 1.94	-5657.08269	0.45
6	Edge OH7 group	0/1	As---OH7: 2.67	-5657.075732	0.65

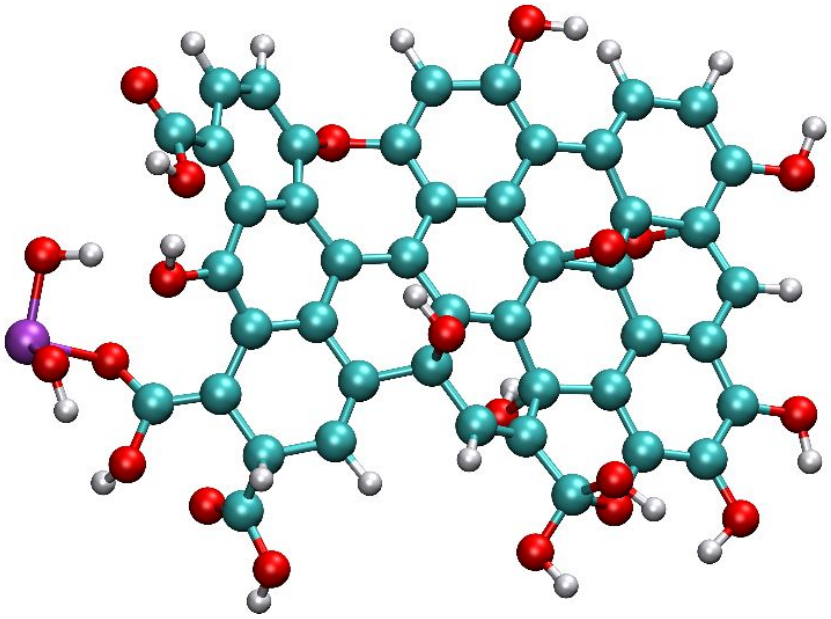


Figure S23. Covalent-bonded As^{3+}-ERGO adduct. The third hydrogen atom of $As(OH)_3$ been lost to the solvent and is not shown. The violet sphere represents the arsenic atom, red - oxygen, cyan - carbon, and white - hydrogen.

(c) Analyte ion-ERGO adducts and binding energies

We further investigated the binding of several metal ions, $[M(H_2O)_6]^{2+}$, where M=Fe, Mn, Cu, and also $H_2PO_4^-$, to ERGO by substituting the $As(OH)_3$ molecule in isomer 2 with these four ions and optimizing their structures using GPAW. The optimized structures of the four adducts are shown in Figures S24 to S27, below. To create these isomers, we selected the structure of isomer 2 as it was the lowest-energy isomer in which $As(OH)_3$ is bound to edge COOH groups, since our experiments indicated structural changes at the COOH group. We also optimized the structure of isomer 2 itself, using GPAW, so that its binding energy could be compared with the other analyte ions. We computed binding energies approximately without basis-set superposition error (BSSE) corrections as E(Binding)=E(Adduct)-E(ERGO)-E(Analyte), and these are listed in Table S5, below.

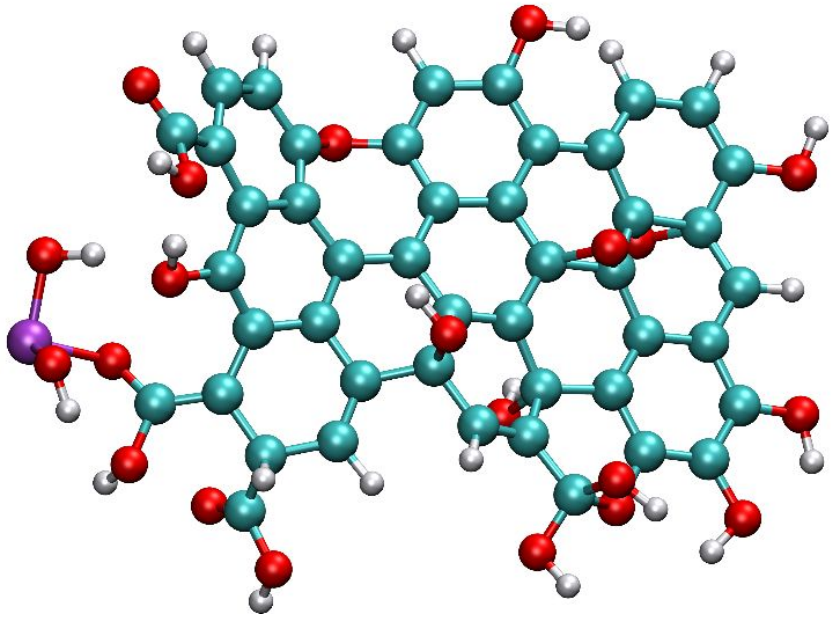


Figure S24. The optimized structure of $Mn(H_2O)_6$-ERGO. The magenta sphere is the manganese atom, red -oxygen, cyan- carbon, and white- hydrogen.

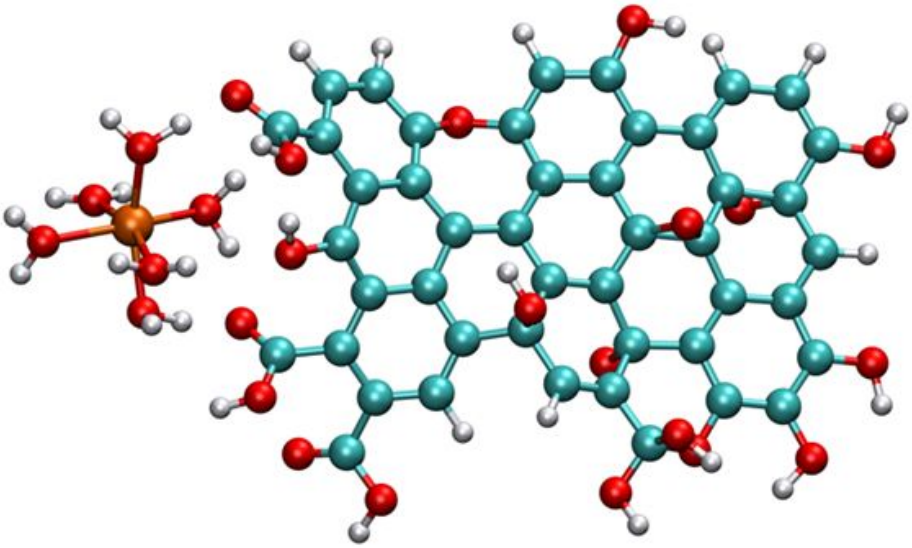


Figure S25. The optimized structure of Fe^{2+}-ERGO, rust-orange sphere is the iron atom; red – oxygen, cyan - carbon and white - hydrogen.

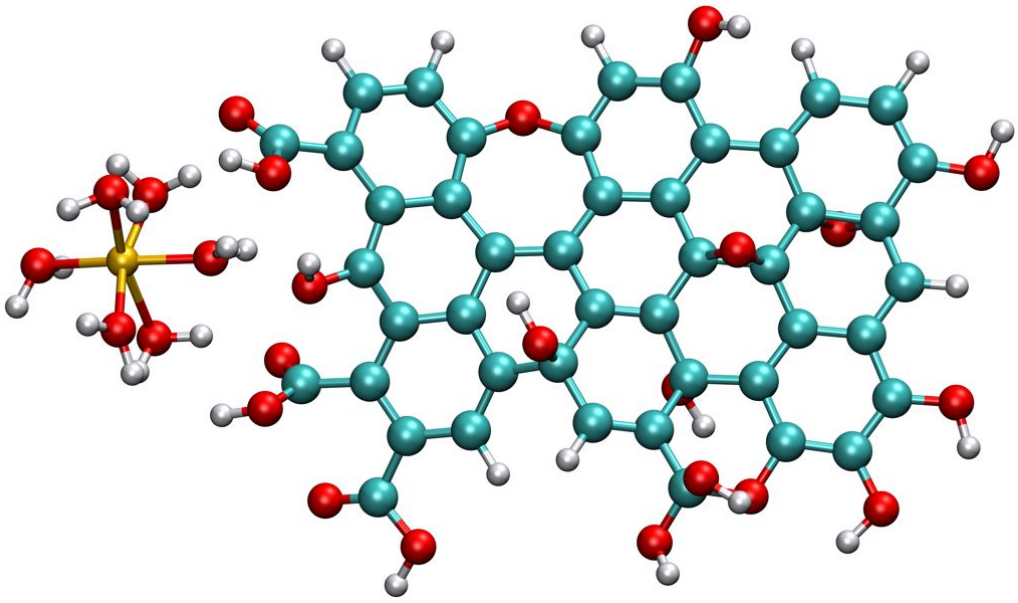


Figure S26. The optimized structure of Cu^{2+}-ERGO, gold sphere is the copper atom; red – oxygen; cyan - carbon and white - hydrogen.

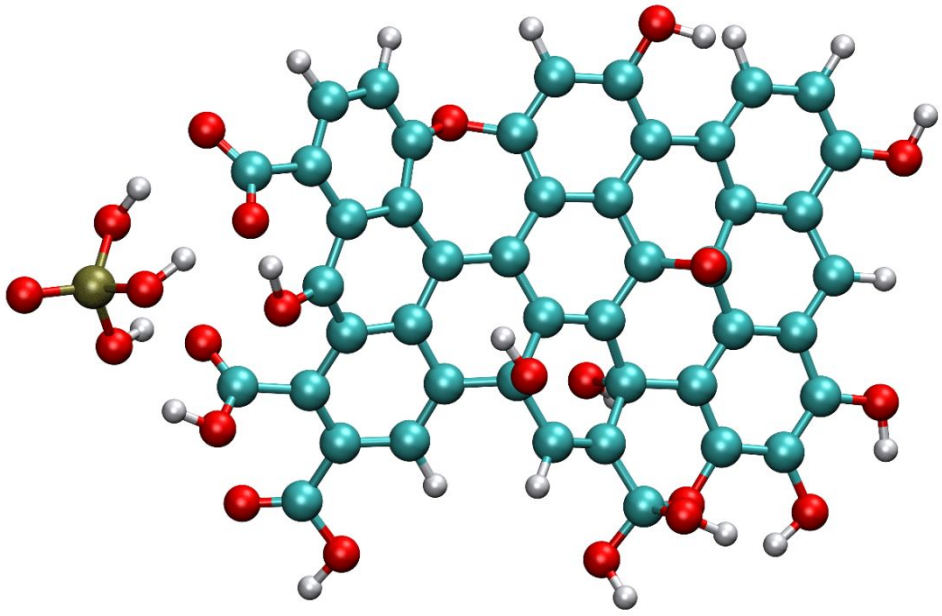


Figure S27. The optimized structure of H_2PO_4 -ERGO adduct. In (c) a small structural change is observed as the H atom of the COOH group has moved its position. Brown spheres are phosphorus atoms; red - oxygen, cyan - carbon and white - hydrogen.

Table S5. The binding energies of selected analyte-ion-ERGO adducts are computed as the difference between the total adduct energy and the sum of the analyte and ERGO energies.

Analyte ion	Species	Analyte total energy/eV	Total adduct energy/eV	Total ERGO energy/eV	Binding energy/eV
Mn^{2+}	$[Mn(H_2O)_6]^{2+}$	-72.110	-661.513	-582.859	-6.54
As^{3+}	$As(OH)_3$	-33.426	-617.064	-582.859	-0.78
Fe^{2+}	$[Fe(H_2O)_6]^{2+}$	-32.927	-620.215	-582.859	-4.43
Cu^{2+}	$[Cu(H_2O)_6]^{2+}$	-66.301	-655.560	-582.859	-6.39
Phosphate	HPO_4^-	-34.465	-625.719	-582.859	-8.39

S24. Methodology of arsenite oxidation mechanism at COOH group of ERGO

We investigated the oxidation mechanism of arsenite with ERGO using universal force field[16] (UFF) simulations based on the starting structure of isomer 2, by reorienting the $As(OH)_3$ and then manually introducing an As-O bond at the ketone O atom followed by a force-field optimization. We then sequentially modified and optimized the geometry to generate a series of intermediate structures, starting from the approach and the covalent binding of $As(OH)_3$ (steps 1 and 2 in Figure S28(b) and (c)) to the regeneration of COOH (steps 3 and 4, in Figure S28(d) and (e)) and final detachment of $HAsO_4^-$ with a regenerated COOH group and conversion of COH to CH (Figure S28 (f)). We also cut out a cluster of atoms from the structure of isomer 2 containing only the arsenite and the COOH1 and OH1 groups and their two neighbouring benzene rings, and performed a similar stepwise structure optimization for the intermediate structures, and these are shown in Figure 5 in the manuscript.

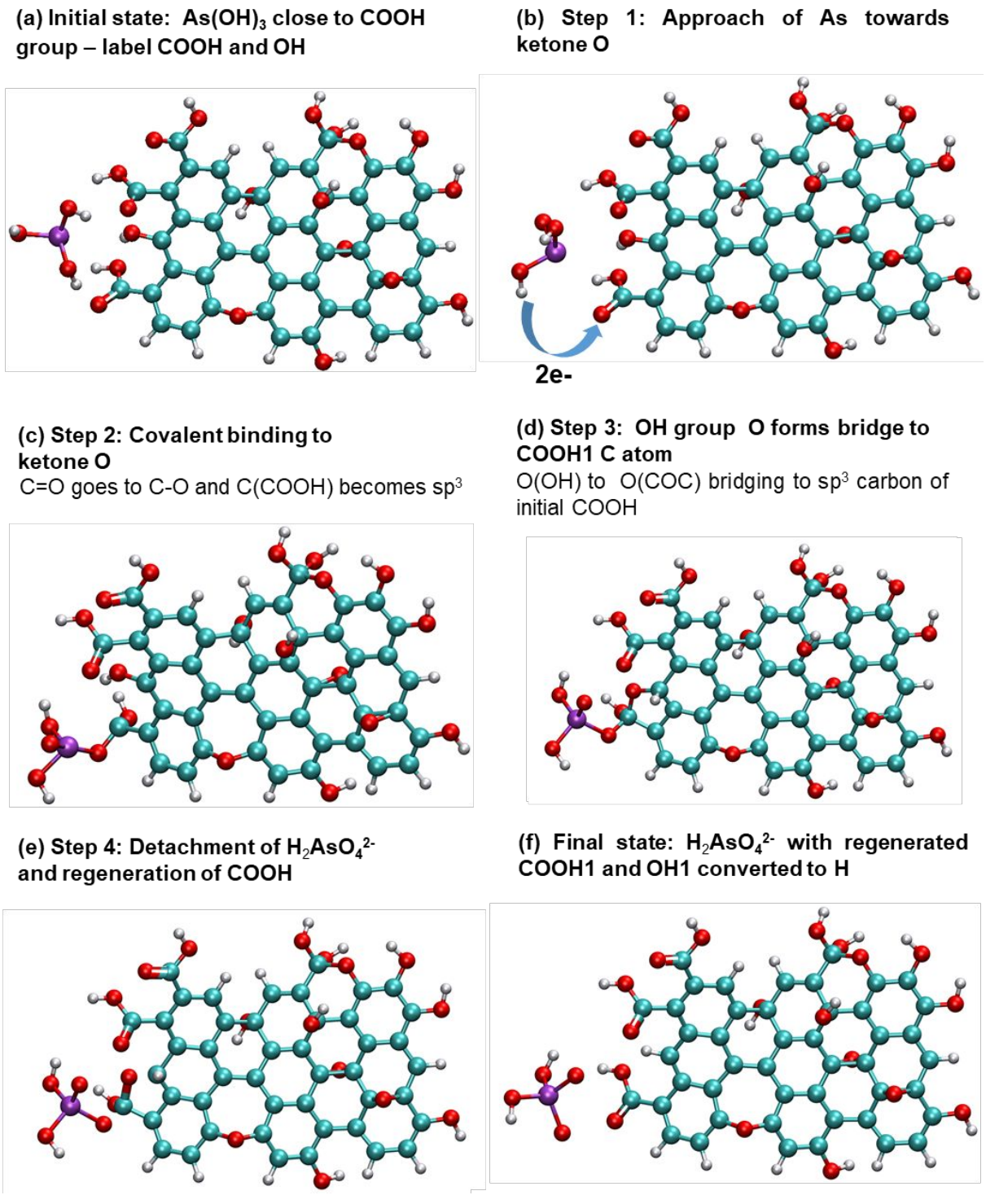


Figure S28. Mechanism of arsenite oxidation, showing the full ERGO sheet. The orientation of the ERGO sheet has been rotated 180° anticlockwise about a horizontal axis in the plane of the page, compared to that in of both Figure 5 and Figure S22 to show the COOH1 group active site clearly. (a) The initial state in the configuration of isomer 2, (b) Step 1, approach of As^{3+} and electron transfer to COOH1 group of ERGO. (c) Step 2, Covalent binding of As to ketone

O to form intermediate As^{5+} species, (d) Step 3, regeneration intermediate step where the oxygen atom of the hydroxyl group binds to the initial COOH1 carbon. (e) Step 4, detachment of $H_2AsO_4^{1-}$. (f) Final state of As^{5+} with COOH group restored with loss of OH oxygen atom from ERGO. Double bonds of C=O are explicitly shown here for clarity in this figure. Atomic colors are violet for arsenic, cyan for carbon, red for oxygen and white for hydrogen.

S25. Methodology of ion-selectivity mechanism

To understand the ion-selectivity mechanism occurring via electron transfer from analyte species to ERGO, we conducted a frontier orbital energy-level analysis of ERGO model and analyte ions. The analyte species considered were, $As(OH)_3$, $HAsO_4$, $[Mn(H_2O)_6]^{2+}$, $[Fe(H_2O)_6]^{2+}$, $[Cu(H_2O)_6]^{2+}$, $H_2PO_4^-$, HPO_4^{2-}, SO_4^{2-}, and NO^{3-}. The values of the HOMO and LUMO levels for all the analyte ions and ERGO models 1, ERGO(6)$^+$ were after geometry optimization in GPAW. The ERGO sheet model 1 was optimized in the +1 charge state, and this model referred was to as ERGO(6)$^+$. We computed the acceptor energy level (LUMO) of the neutral and positively charged charge states of ERGO models 1, 2 and 3, and the results are shown in Table S6 below. We also computed the donor level, which is the HOMO of each analyte species, and results are shown graphically in Figure 6 in the manuscript and tabulated below in Table S7.

Table S6. LUMO acceptor level and HOMO energies for the neutral and positively charged (denoted by affix "+" to the model number) ERGO models 1, 2 and 3 and their band gaps. The neutral model 3 has an unpaired spin, hence, the band gap, E_{gap}, is slightly larger than in the other models.

ERGO model	**Model type and oxygen-carbon ratio**	**Charge**	**LUMO/eV**	**HOMO/eV**	E_{gap}**/eV**
Model 1 (ERGO)	rectangular sheet N_{Oxygen}/N_{Carbon}=0.6	0	-3.71	-3.89	0.18
Model 1^+ (ERGO$^+$ or ERGO6)	rectangular sheet, ERGO$^+$ N_{Oxygen}/N_{Carbon}=0.6	+1	-6.36	-6.53	0.17
Model 2	rectangular sheet N_{Oxygen}/N_{Carbon}=0.3	0	-3.71	-3.99	0.28
Model 2^+	rectangular sheet N_{Oxygen}/N_{Carbon}=0.3	+1	-5.88	-5.99	0.11
Model 3	hexagonal flake N_{Oxygen}/N_{Carbon}=0.3	0	-3.51	-4.21	0.7*
Model 3^+	hexagonal flake N_{Oxygen}/N_{Carbon}=0.3	+1	-6.25	-6.43	0.18

Table S7. Analyte ion donor-level (HOMO) energies and donor-acceptor energy level differences, ΔE_{DA}= E_{HOMO}(Analyte)-E_{LUMO}(ERGO(6)$^{+}$), where the acceptor level energy is E_{LUMO}(ERGO(6)$^{+}$)=-6.36 eV. ERGO^{+} is the optimized geometry of model 1 (ERGO) in the positive charge state.

Analyte ion	**Species**	**HOMO energy (E_{HOMO}) /eV**	**ΔE_{DA}/eV**
As^{3+}	$As(OH)_3$	-6.35	0.01
Mn^{2+}	$[Mn(H_2O)_6]^{2+}$	-3.52	2.84
Fe^{2+}	$[Fe(H_2O)_6]^{2+}$	-10.33	-3.97
Cu^{2+}	$[Cu(H_2O)]_6^{2+}$	-14.56	-14.56
Phosphate	HPO_4^{-}	-0.5	5.86
As^{5+}	$HAsO_4^{2-}$	6.17	12.53
Sulfate	SO_4^{2-}	6.83	13.18
Phosphate	$H_2(PO_4)^{2-}$	6.90	13.26
Nitrate	NO_3^{-}	11.54	17.54

S26. Atomic coordinates of the optimized structures

All coordinates in XYZ file format in Angstroms and the optimization method is indicated**.**

(a) Non-covalently bound $As(OH)_3$-ERGO adduct isomers 1 to 6

All geometries were optimized using NWChem at B3LYP/6-31G* level

Isomer 1:

```
92

C          -0.29887       -0.37517        0.97869
C           0.84738       -1.07854        0.79955
C           2.14422       -0.38256        0.76262
C           3.34529       -1.03843        0.26396
C           4.52101       -0.30829        0.11645
C           5.78598       -0.95585       -0.08089
C           6.95294       -0.21236       -0.17665
C          -0.34383        1.06202        0.93450
C           0.74036        1.86098        0.65247
C           2.05212        1.16979        0.68508
C           3.22303        1.83826        0.07991
C           4.51646        1.13065        0.15510
C           5.68672        1.86534        0.06823
C           6.90805        1.18686       -0.06530
C          -0.43780        4.10060        0.61844
C           0.74755        3.33839        0.52427
C           1.96453        4.07790        0.36241
C           3.18781        3.34417        0.01469
C           4.43902        4.07915        0.01201
C           5.73192        3.35311        0.32146
C           6.92081        3.97891       -0.37178
C          -0.37349        5.47067        0.86051
C           0.83813        6.12422        0.84356
C           2.03901        5.50347        0.43337
C           3.25524        6.25026        0.09728
C           4.42137        5.44882       -0.12305
C           5.64843        6.09915       -0.73335
C           6.87347        5.23669       -0.80546
C          -0.04385        9.30327        0.26971
C           1.02051        8.44604        0.43896
C           2.31727        8.68130       -0.09554
C           3.38699        7.71006       -0.03550
C           4.70959        8.28389       -0.09493
C           5.87535        7.46834       -0.15534
C           7.11590        7.98266        0.11824
C           0.12766       10.47048       -0.48082
C           1.38012       10.84690       -0.93094
C           2.53930       10.02842       -0.59834
C           3.85460       10.53738       -0.51502
C           4.92201        9.70026       -0.09698
C           6.21908       10.23165        0.21017
C           7.28721        9.36412        0.39154
O           0.93007       -2.42624        0.63203
H           3.32912       -2.10683        0.09272
```

O	5.82678	-2.30575	-0.14404
C	1.33579	12.05872	-1.78316
O	4.17432	11.82440	-0.75017
C	6.40280	11.71332	0.46677
H	-1.24139	-0.90771	1.08939
H	-1.30935	1.48989	1.16614
O	8.13165	-0.88976	-0.33632
O	8.10933	1.81679	-0.02382
O	-1.69986	3.58218	0.53662
H	-1.28280	6.01749	1.08111
C	8.22838	3.20924	-0.52973
H	7.72659	5.67240	-1.31618
H	-0.99397	9.05582	0.72884
H	-0.71643	11.11028	-0.71008
H	7.98826	7.34246	0.12889
C	8.61799	9.86856	0.83184
H	0.03437	-2.79301	0.55251
H	6.75894	-2.57387	-0.24550
O	0.42646	12.86024	-1.83558
O	2.41122	12.19611	-2.64250
H	2.22951	13.00394	-3.16021
H	3.64117	12.17558	-1.49266
O	6.02963	12.26052	1.47542
O	7.03768	12.33364	-0.54810
H	7.18072	13.25197	-0.24800
H	8.85131	-0.23264	-0.33723
H	7.94365	2.89344	-2.42881
H	-1.66711	2.77734	-0.00577
O	8.68493	3.12781	-1.83108
O	9.21063	3.78925	0.23225
H	10.06259	3.46877	-0.11492
O	8.85981	10.99383	1.21659
O	9.57937	8.90979	0.77794
H	10.39261	9.33979	1.09907
O	2.36495	0.50295	1.89913
O	2.96818	2.44664	-1.19513
O	0.82846	7.41592	1.32149
O	5.35653	6.30378	-2.16256
H	4.49210	6.74303	-2.23174
O	5.87847	3.55127	1.76347
H	6.65955	3.04589	2.05391
As	5.88367	3.14794	-3.91989
O	4.17313	3.50280	-3.51833
O	6.28088	4.81701	-4.43434
O	5.77187	2.45886	-5.56711
H	5.38770	3.13682	-6.15432
H	5.87229	5.41925	-3.77256
H	3.86910	2.96373	-2.76352

Isomer 2:

92

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024

C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975
C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811
C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793
C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	3.07105	10.59968	-0.71189
C	4.20242	9.92929	-0.20956
C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	0.56704	12.01273	-1.76324
O	3.19949	11.92668	-0.99920
C	5.23819	12.12107	0.51335
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495
C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	-0.16729	12.92746	-1.43887

O	1.36562	12.13187	-2.87923
H	1.20382	13.03108	-3.22770
H	2.86943	12.08916	-1.90491
O	4.57238	12.50871	1.45824
O	5.83312	12.91015	-0.37822
H	5.53004	13.83466	-0.18694
H	9.54686	0.69774	0.45476
H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600
H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	2.72239	15.14463	-0.78007
O	1.50043	14.69819	0.46497
O	4.14151	15.00383	0.34666
O	2.66056	16.93431	-0.73496
H	2.87140	17.22456	0.17164
H	3.97295	14.28940	1.00560
H	0.78616	14.20087	0.02304

Isomer 3:

92

C	-0.03892	-0.14621	0.03346
C	1.08646	-0.73953	-0.45478
C	2.38718	-0.06532	-0.29890
C	3.59251	-0.53368	-1.00362
C	4.78108	0.18198	-0.89703
C	6.04844	-0.35725	-1.30104
C	7.22053	0.33908	-1.02894
C	-0.06672	1.22770	0.46262
C	1.00872	2.08252	0.36092
C	2.30790	1.40890	0.13780
C	3.49050	2.22887	-0.24903
C	4.78371	1.50269	-0.31944
C	5.95217	2.17287	-0.02441
C	7.17051	1.56159	-0.33860
C	-0.18376	4.28621	0.76629
C	1.00750	3.55249	0.53972
C	2.21257	4.31276	0.47817
C	3.46356	3.64407	0.08176
C	4.71669	4.34781	0.32697
C	5.94837	3.53960	0.65032
C	7.21749	4.25715	0.27725
C	-0.11860	5.61793	1.16649
C	1.08779	6.28678	1.16950
C	2.28233	5.72014	0.68478

C	3.51657	6.49406	0.45326
C	4.71651	5.71499	0.27926
C	5.99174	6.38749	-0.19096
C	7.23321	5.54434	-0.06431
C	0.12231	9.39709	0.64420
C	1.22660	8.59242	0.83185
C	2.50679	8.87690	0.29255
C	3.61722	7.94268	0.36080
C	4.92606	8.56431	0.31786
C	6.12190	7.78957	0.34852
C	7.32910	8.37507	0.64045
C	0.24747	10.56382	-0.11279
C	1.48346	10.99036	-0.57276
C	2.67807	10.22366	-0.22910
C	3.98062	10.77005	-0.17731
C	5.07920	9.98937	0.26573
C	6.34334	10.58544	0.59006
C	7.44209	9.77108	0.83593
O	1.13186	-1.94187	-1.08872
H	3.55630	-1.47624	-1.53464
O	6.08388	-1.56037	-1.93171
C	1.39076	12.17410	-1.46205
O	4.25486	12.06184	-0.44576
C	6.46259	12.07806	0.80423
H	-0.98180	-0.68999	0.02077
H	-1.01188	1.56378	0.87071
O	8.40711	-0.24824	-1.39017
O	8.38780	2.08360	0.03277
O	-1.44550	3.77382	0.63786
H	-1.02618	6.13275	1.46026
C	8.50200	3.47460	0.42853
H	8.16245	6.00165	-0.38113
H	-0.81624	9.11356	1.10760
H	-0.62353	11.16671	-0.34521
H	8.23166	7.78118	0.71696
C	8.74290	10.34107	1.28711
H	0.22554	-2.25951	-1.23117
H	7.01410	-1.78352	-2.09977
O	0.39003	12.88232	-1.53281
O	2.46010	12.37448	-2.26631
H	2.32524	13.16900	-2.87710
H	3.66868	12.39461	-1.16380
O	6.03594	12.64389	1.77913
O	7.12436	12.68475	-0.20341
H	7.21897	13.62056	0.06847
H	9.12194	0.38289	-1.20480
H	10.32407	4.00306	0.22201
H	-1.41632	3.04589	-0.00703
O	9.51329	4.04914	-0.31720
O	8.86003	3.40423	1.80197
H	9.12539	2.47823	1.96854
O	8.93738	11.47887	1.66566
O	9.74520	9.42208	1.26318
H	10.53440	9.88864	1.60194
O	2.63444	0.37485	1.06502
O	3.29867	3.12060	-1.35541
O	1.07381	7.55060	1.71687
O	5.87975	6.51411	-1.64290

H	5.10756	7.07949	-1.82645
O	5.89259	3.36001	2.09560
H	6.77555	3.06328	2.38898
As	0.67066	14.87605	-4.79550
O	0.47653	13.14214	-5.32954
O	2.30046	14.44490	-4.00897
O	-0.25210	14.97127	-3.29870
H	-0.07306	14.22692	-2.67414
H	2.85070	14.10163	-4.74095
H	-0.30446	13.04351	-5.89644

Isomer 4:

92

C	-0.25497	0.18117	0.49210
C	0.88416	-0.55035	0.59324
C	2.19241	0.11743	0.68649
C	3.43080	-0.60746	0.45987
C	4.63649	0.08655	0.37625
C	5.89811	-0.59436	0.41739
C	7.08737	0.10919	0.30500
C	-0.25202	1.60173	0.27272
C	0.88583	2.35226	0.08305
C	2.15703	1.66252	0.40728
C	3.42110	2.23144	-0.09264
C	4.66783	1.51569	0.21440
C	5.86525	2.21237	0.12099
C	7.06700	1.50575	0.17045
C	-0.21967	4.58433	-0.35185
C	0.94761	3.80865	-0.17719
C	2.18581	4.52630	-0.17611
C	3.42651	3.75155	-0.31374
C	4.68516	4.46161	-0.19883
C	5.93329	3.71761	0.22670
C	7.17798	4.26894	-0.42749
C	-0.17371	5.96316	-0.16991
C	1.02968	6.60242	0.03260
C	2.27805	5.95519	-0.12163
C	3.55567	6.66461	-0.24064
C	4.71990	5.82820	-0.32602
C	6.02969	6.45756	-0.77311
C	7.22459	5.54439	-0.79399
C	0.40614	9.87542	-0.60562
C	1.36993	8.95080	-0.26598
C	2.76373	9.14592	-0.48179
C	3.76481	8.12309	-0.27855
C	5.10740	8.63080	-0.09037
C	6.23658	7.76329	-0.05937
C	7.44853	8.18997	0.41609
C	0.77760	11.08365	-1.20069
C	2.11506	11.41779	-1.32732
C	3.13504	10.51194	-0.81525
C	4.41779	10.94912	-0.43174
C	5.37468	10.02956	0.06102
C	6.63578	10.46593	0.59225
C	7.63735	9.53480	0.82612
O	0.94598	-1.91014	0.59928

H	3.40620	-1.68738	0.40178
O	5.90876	-1.93927	0.58285
C	2.34329	12.65625	-2.08794
O	4.77951	12.25542	-0.44876
C	6.83766	11.90858	1.00465
H	-1.21837	-0.32502	0.50775
H	-1.22912	2.06564	0.28983
O	8.26009	-0.60566	0.35156
O	8.29016	2.13639	0.13437
O	-1.45007	4.06820	-0.64047
H	-1.09676	6.53079	-0.14798
C	8.35369	3.34713	-0.68240
H	8.11509	5.96028	-1.25289
H	-0.63351	9.64599	-0.40297
H	0.00756	11.75870	-1.55631
H	8.28424	7.50490	0.47814
C	8.92374	9.93676	1.46086
H	0.06207	-2.26874	0.41758
H	6.83802	-2.22593	0.63404
O	1.53946	13.55981	-2.27029
O	3.55988	12.72998	-2.72580
H	3.54288	13.57007	-3.22431
H	4.56682	12.63914	-1.32234
O	6.36558	12.38236	2.00901
O	7.61590	12.58761	0.13560
H	7.74974	13.46715	0.53714
H	8.99143	0.02878	0.43785
H	8.71918	2.05283	-2.07242
H	-1.32833	3.21485	-1.08718
O	8.32962	2.94309	-2.02450
O	9.53831	3.97247	-0.34570
H	10.23458	3.60769	-0.91680
O	9.15893	11.01086	1.97434
O	9.85080	8.94375	1.42880
H	10.63287	9.30492	1.88543
O	2.25657	1.13983	1.72198
O	3.37206	2.76824	-1.41293
O	0.92254	7.89810	0.48196
O	5.89904	6.75361	-2.19650
H	5.11504	7.31798	-2.30336
O	5.99082	4.03248	1.65813
H	6.65678	3.44474	2.05853
As	-2.04404	14.12244	-2.51335
O	-0.87087	14.68045	-1.26478
O	-3.38892	14.97620	-1.67617
O	-2.32471	12.42360	-1.92034
H	-2.59941	12.46659	-0.98612
H	-3.00263	15.36303	-0.86865
H	0.02313	14.34599	-1.48657

Isomer 5:

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92
C          0.70027       -0.70103        1.10266
C          1.93033       -1.25851        0.95842
C          3.12506       -0.40661        0.84511
C          4.39046       -0.93763        0.36695
C          5.45156       -0.07215        0.10590
C          6.78415       -0.56158       -0.09902
C          7.82543        0.31247       -0.37415
C          0.46276        0.70866        0.95578
C          1.42748        1.62335        0.59753
C          2.82229        1.12021        0.65059
C          3.88032        1.87787       -0.04371
C          5.25199        1.34895        0.00735
C          6.30401        2.21671       -0.25830
C          7.58501        1.69333       -0.43956
C         -0.03689        3.69175        0.49736
C          1.23560        3.08061        0.39950
C          2.34055        3.96371        0.18775
C          3.63087        3.38077       -0.20467
C          4.77646        4.26912       -0.29187
C          6.17362        3.71434       -0.10660
C          7.19764        4.45243       -0.93873
C         -0.13530        5.06202        0.73597
C          0.99087        5.84714        0.71170
C          2.24414        5.38792        0.27470
C          3.34906        6.27585       -0.08503
C          4.58899        5.62482       -0.39498
C          5.68355        6.43615       -1.07155
C          6.98862        5.71990       -1.28910
C         -0.29570        8.87029        0.19756
C          0.87694        8.17477        0.34975
C          2.12399        8.58259       -0.18091
C          3.30448        7.74522       -0.17602
C          4.54927        8.47211       -0.27466
C          5.79427        7.79667       -0.43737
C          6.98203        8.44248       -0.21071
C         -0.28489       10.07027       -0.52670
C          0.90989       10.63164       -0.95046
C          2.16857        9.96394       -0.63561
C          3.41459       10.62653       -0.54552
C          4.59329        9.90214       -0.21715
C          5.83868       10.56601        0.05084
C          7.00845        9.82157        0.12281
O          2.18341       -2.59339        0.88744
H          4.50889       -2.00788        0.26601
O          7.01279       -1.89373       -0.00061
C          0.70494       11.84734       -1.77777
O          3.56314       11.95878       -0.68938
C          5.86807       12.04276        0.38796
H         -0.16184       -1.34361        1.26928
H         -0.54834        1.02190        1.17420
O          9.07839       -0.22244       -0.55484
O          8.68361        2.49782       -0.64238
O         -1.22441        3.01943        0.43788
H         -1.09849        5.49292        0.98388
C          8.44416        3.72369       -1.40237
```

H	7.71931	6.26029	-1.88084
H	-1.20446	8.48818	0.64827
H	-1.20965	10.59078	-0.74659
H	7.92060	7.90976	-0.28853
C	8.29811	10.46001	0.50665
H	1.34143	-3.07487	0.85426
H	7.96769	-2.04236	-0.11897
O	-0.32062	12.49347	-1.84570
O	1.77592	12.18259	-2.58182
H	1.48757	12.96807	-3.08389
H	2.98432	12.29196	-1.40402
O	5.49189	12.48395	1.44901
O	6.36802	12.79042	-0.61822
H	6.44861	13.69482	-0.25993
H	9.71643	0.51057	-0.55716
H	8.79734	2.54587	-2.89504
H	-1.09202	2.20478	-0.07185
O	8.27555	3.35257	-2.74257
O	9.55960	4.51537	-1.21095
H	10.22861	4.24608	-1.86153
O	8.43173	11.58054	0.94731
O	9.36125	9.63378	0.32954
H	10.13639	10.13997	0.63342
O	3.23588	0.59150	1.90075
O	3.51277	2.41333	-1.31491
O	0.83779	7.12074	1.22240
O	5.26247	6.63763	-2.45708
H	4.40131	7.08594	-2.43520
O	6.42584	4.01592	1.30593
H	7.22259	3.52308	1.57150
As	-0.12655	9.17742	4.48386
O	0.55629	7.53869	4.08006
O	0.33546	8.98328	6.22570
O	-1.88344	8.81184	4.64278
H	-1.99299	8.12530	5.33732
H	0.84675	8.15300	6.27285
H	0.64085	7.44656	3.10704

Isomer 6:

92

C	0.50719	-0.54540	0.53673
C	1.73582	-1.12542	0.50808
C	2.95176	-0.29689	0.56393
C	4.25313	-0.83761	0.21097
C	5.35158	0.01340	0.09796
C	6.68982	-0.49525	0.01510
C	7.76832	0.36429	-0.12343
C	0.31761	0.87410	0.41625
C	1.33227	1.77878	0.19870
C	2.70056	1.24455	0.39119
C	3.84255	2.00267	-0.14982
C	5.18900	1.44099	0.02974
C	6.27841	2.29448	-0.09148
C	7.56080	1.75159	-0.16237
C	-0.07784	3.86645	-0.02161

C	1.18843	3.24271	0.02966
C	2.32158	4.11636	-0.00535
C	3.63804	3.52084	-0.27136
C	4.80083	4.38285	-0.19080
C	6.15983	3.78684	0.10956
C	7.27697	4.53502	-0.57914
C	-0.19549	5.22886	0.23781
C	0.92725	6.01029	0.39885
C	2.23403	5.54054	0.12929
C	3.39881	6.41681	-0.02449
C	4.65186	5.74750	-0.23180
C	5.83760	6.56933	-0.71155
C	7.13524	5.82456	-0.86287
C	-0.15164	9.18857	-0.04473
C	0.94399	8.39183	0.20240
C	2.28463	8.78387	-0.07475
C	3.41718	7.89116	0.01542
C	4.69125	8.55853	0.16343
C	5.92198	7.84634	0.07581
C	7.09582	8.39937	0.51425
C	0.02901	10.45922	-0.59882
C	1.29805	10.98171	-0.76599
C	2.45579	10.20263	-0.34944
C	3.69391	10.78378	0.00183
C	4.78567	9.96893	0.39108
C	6.00990	10.53402	0.88605
C	7.13464	9.73011	1.00542
O	1.97602	-2.46070	0.41002
H	4.36259	-1.90636	0.08596
O	6.88545	-1.83521	0.08923
C	1.28901	12.29050	-1.45900
O	3.89464	12.11839	0.04998
C	6.05068	11.96247	1.38547
H	-0.38438	-1.16656	0.58121
H	-0.70677	1.20198	0.53422
O	9.02429	-0.19067	-0.19707
O	8.68937	2.53929	-0.22955
O	-1.25020	3.20975	-0.26100
H	-1.17981	5.66727	0.35480
C	8.54162	3.79314	-0.96314
H	7.93243	6.38016	-1.34499
H	-1.13656	8.81345	0.20885
H	-0.82294	11.06232	-0.89139
H	8.01487	7.82816	0.48477
C	8.39254	10.25663	1.60382
H	1.14280	-2.93248	0.23010
H	7.84552	-1.99628	0.06594
O	0.33088	13.02462	-1.58046
O	2.47796	12.61844	-2.08633
H	2.30872	13.47411	-2.52552
H	3.50354	12.54202	-0.74185
O	5.57518	12.31004	2.43881
O	6.69222	12.78761	0.53154
H	6.73377	13.65093	0.98490
H	9.67110	0.52955	-0.10964
H	8.96354	2.63427	-2.45315
H	-1.05092	2.39116	-0.74349
O	8.48251	3.47042	-2.32705

```
O          9.65488        4.55090       -0.65511
H         10.35988        4.29894       -1.27458
O          8.51616       11.31360        2.18753
O          9.43874        9.40292        1.44800
H         10.19201        9.83403        1.89217
O          2.96131        0.66384        1.65874
O          3.63115        2.60204       -1.42680
O          0.68959        7.25913        0.92540
O          5.57652        6.93037       -2.10193
H          4.71758        7.38463       -2.12477
O          6.27351        4.02337        1.55297
H          7.02176        3.48963        1.87616
As        -1.12946       -4.29461       -0.21634
O         -1.75257       -5.24708       -1.62528
O         -1.80858       -5.46801        0.95178
O         -2.34400       -2.98598       -0.05192
H         -3.21075       -3.36105        0.18958
H         -2.16828       -6.21703        0.44223
H         -1.35151       -4.93484       -2.45359
```

(b) Covalently bonded As(OH)$_3$-ERGO model

Geometry was optimized using GPAW at GGA-LCAO basis level and atomic positions are in Angstroms.

```
91

C 12.1677668600 9.0251289400 17.6885076800
C 13.4053704300 8.4516831400 17.5057732800
C 14.6181720900 9.2928537800 17.4028667800
C 15.8915886100 8.7475741500 16.9321399400
C 16.9697034700 9.6179303000 16.6817014000
C 18.3093981300 9.1237747800 16.4810008700
C 19.3644328400 10.0028629000 16.2004435700
C 11.9461727900 10.4438310100 17.5767685600
C 12.9327092600 11.3571578800 17.2031791400
C 14.3260427100 10.8283557100 17.2256398000
C 15.3940655400 11.5856903100 16.5230990800
C 16.7729064100 11.0499569500 16.5725701500
C 17.8390473400 11.9214224800 16.3123835300
C 19.1351426300 11.3940772900 16.1317715900
C 11.4896357600 13.4473613200 17.1958160300
C 12.7636087000 12.8178706100 17.0499705000
C 13.8806801100 13.6964030800 16.8286807000
C 15.1576754000 13.1013069800 16.3959467400
C 16.3178191900 13.9790751300 16.2869490600
C 17.7209726500 13.4263117900 16.4326425700
C 18.7072803200 14.1360498000 15.5320096600
C 11.4173629200 14.8161597200 17.4937948000
C 12.5614780300 15.6066501900 17.4512582400
C 13.8046397600 15.1323401400 16.9401484400
C 14.9047383500 16.0104020200 16.5200405200
C 16.1258995700 15.3472927800 16.1477764400
C 17.1709294600 16.1234170100 15.3516769500
C 18.4523610300 15.3735536200 15.0765267900
C 11.2889060900 18.7130875500 17.0789533000
C 12.4625988800 17.9483406000 17.1324971900
```

C 13.6698357600 18.3208555900 16.4805460900
C 14.8750223900 17.4853279300 16.4239107900
C 16.1035184700 18.2010495300 16.2339357100
C 17.3620507000 17.5140237900 15.9208814700
C 18.5572181300 18.1572265200 15.8402429300
C 11.2626066200 19.8737108800 16.2982261200
C 12.4362344900 20.3554163800 15.6887561400
C 13.6960477300 19.6649645500 15.9240891300
C 14.9730872200 20.3140675500 15.8461055200
C 16.1561768700 19.6408269100 16.2161939600
C 17.4318964800 20.3386058200 16.4190017200
C 18.7412365800 19.6255347000 16.1216404000
O 13.6359547800 7.1042406900 17.3859506600
H 16.0163109500 7.6546196700 16.8483356700
O 18.5588037200 7.7821385600 16.5870308500
C 12.1787627700 21.5115747400 14.7951607900
O 15.0816603900 21.6428368100 15.4900076600
C 17.5156667600 21.5749245200 17.0263850500
H 11.2922469400 8.3665124600 17.8625970500
H 10.9286176100 10.7766363900 17.8501212600
O 20.6157641500 9.4384893400 16.0130335200
O 20.2407877100 12.1971167400 15.9166591500
O 10.2880075600 12.7881277700 17.1134193000
H 10.4497914300 15.2667167400 17.7780985600
C 19.9682840000 13.4016743000 15.0995662400
H 19.1645179400 15.8596610000 14.3830514700
H 10.4017995600 18.3584771800 17.6351238500
H 10.3287465100 20.4458540200 16.1414529400
H 19.4665925100 17.6021005600 15.5414200100
C 19.7805251000 19.7288276900 17.2628634600
H 12.7590249000 6.6524983800 17.3644108800
H 19.5327253000 7.6735925000 16.4440099300
O 11.1662389400 22.2100323700 14.7791981700
O 13.1697212500 21.7267465200 13.8093589400
H 12.8283598900 22.5181802700 13.3163133900
H 14.4621225600 21.7948116800 14.7059199500
O 16.5637208200 22.1331211900 17.7832772600
O 18.7141934500 22.2850630100 16.9240759800
H 21.2487228600 10.1951762600 15.9424466800
H 20.4342863600 12.2448462500 13.6138661700
H 10.4673779400 11.9237782000 16.6674316400
O 19.7858072900 12.9769648500 13.7636899200
O 21.0746131000 14.2360681000 15.2713306000
H 21.8194431700 13.8231873200 14.7700181100
O 19.5804533700 20.0220337200 18.4366844600
O 21.0199911800 19.3936007600 16.7750031600
H 21.6137924300 19.4113648600 17.5682545100
O 14.7450414100 10.2839854400 18.4926202400
O 15.0112527600 12.1501443300 15.2465777000
O 12.4232598100 16.8729546300 18.0048822000
O 16.6160886700 16.2833837800 13.9992672300
H 15.8120809000 16.8471087600 14.1150386500
O 18.1781063900 13.7782706900 17.7960783000
H 17.6136155500 13.2377497000 18.4035666600
As 16.0802972400 23.9969795300 17.5436022500
O 14.3735845900 23.6419806700 17.1942134000
O 16.6412965400 24.2032287100 15.8218255600
H 17.6024087500 23.9501117800 15.8155410200

```
H 14.4478124600 22.8606232300 16.5539830400
H 19.2645631700 20.0669711800 15.2314524200
H 19.0113216100 22.4730159100 17.8509776900
```

(c)Analyte-ERGO adducts for selected analyte ions

All geometries were optimized using GPAW with a DZP basis set and a GGA-PBE functional.

ERGO-$[Mn(H_2O)_6]^{2+}$

```
104

C        0.4082500000     -0.8237100000      0.5226400000
C        1.6500600000     -1.3054500000      0.6008700000
C        2.8002500000     -0.3857400000      0.6118100000
C        4.1906000000     -0.9411500000      0.4084900000
C        5.3157400000      0.0261200000      0.3237300000
C        6.6298100000     -0.4401400000      0.4030000000
C        7.6843400000      0.4651100000      0.3366800000
C        0.1425200000      0.6168900000      0.3737600000
C        1.1397900000      1.5305700000      0.1946300000
C        2.5384900000      1.0333700000      0.3545700000
C        3.6732400000      1.9085200000      0.0216300000
C        5.0652800000      1.4031800000      0.1792600000
C        6.1246800000      2.3021600000      0.1825200000
C        7.4273300000      1.8267700000      0.2310500000
C       -0.3386600000      3.5752700000     -0.0880700000
C        0.9369300000      3.0233200000     -0.0043200000
C        2.0606400000      3.9215400000     -0.0437400000
C        3.4271400000      3.3423600000     -0.1388300000
C        4.5774000000      4.2281400000     -0.3303400000
C        5.8996800000      3.7970600000      0.3159800000
C        7.0254900000      4.5554800000     -0.3419400000
C       -0.4955200000      4.8944100000      0.1330900000
C        0.5528700000      5.7753500000      0.2293400000
C        1.8896700000      5.3558700000      0.0047300000
C        3.0316900000      6.2488500000     -0.1426500000
C        4.3005000000      5.6810400000     -0.3007600000
C        5.4739600000      6.4917800000     -0.7892300000
C        6.8291800000      5.8045800000     -0.7916300000
C       -0.6687100000      8.9219100000     -0.1273000000
C        0.4381100000      8.1034600000      0.0181300000
C        1.7326900000      8.5227200000     -0.2829400000
C        2.9469100000      7.7097500000     -0.2323000000
C        4.1922400000      8.4428400000     -0.1796400000
C        5.4411100000      7.8251000000     -0.1663800000
C        6.6074800000      8.5260600000      0.2100500000
C       -0.5903700000     10.1298300000     -0.7284300000
C        0.6404100000     10.6294200000     -1.0963700000
C        1.8421300000      9.8695200000     -0.7297600000
C        3.1264300000     10.4155000000     -0.6966500000
C        4.1993000000      9.7983900000     -0.1479100000
```

C	5.3497500000	10.5326300000	0.2474700000
C	6.5438700000	9.8802400000	0.5111200000
O	1.8756800000	-2.6312300000	0.6255100000
H	4.3697100000	-2.0315300000	0.4362000000
O	6.8818900000	-1.7551900000	0.5504100000
C	0.5509300000	11.9213400000	-1.8169400000
O	3.4494100000	11.4751200000	-1.3434200000
C	5.2679400000	11.9606300000	0.4840300000
H	-0.4337100000	-1.4998300000	0.5221800000
H	-0.8920800000	0.8507800000	0.4017500000
O	8.9488200000	0.0206900000	0.3966900000
O	8.4435800000	2.7044800000	0.1729000000
O	-1.4363000000	2.8746100000	-0.4149000000
H	-1.5072500000	5.3031000000	0.1473400000
C	8.3527800000	3.8563300000	-0.5909100000
H	7.6508100000	6.3271000000	-1.2758800000
H	-1.6465200000	8.5308500000	0.1362500000
H	-1.5116200000	10.6753000000	-0.9346000000
H	7.5562000000	8.0276800000	0.2722000000
C	7.7468300000	10.5849400000	1.0416500000
H	1.1648600000	-3.2566300000	0.4473400000
H	7.7907300000	-2.0776700000	0.6197800000
O	-0.2713200000	12.7630800000	-1.4750400000
O	1.1631500000	12.0601100000	-3.0049600000
H	1.0993100000	12.8984500000	-3.4819900000
H	3.2690000000	11.4925100000	-2.2768700000
O	4.6988000000	12.3593700000	1.4933000000
O	5.9113500000	12.7940500000	-0.3668200000
H	5.9468200000	13.7495000000	-0.2818000000
H	9.6868500000	0.6390400000	0.4280600000
H	8.9758700000	2.7064500000	-2.0602100000
H	-1.3673300000	2.1352200000	-1.0371200000
O	8.4128500000	3.5235800000	-1.9656200000
O	9.4091300000	4.7087300000	-0.2464400000
H	10.1921400000	4.4397500000	-0.7923800000
O	7.6868900000	11.7274600000	1.4804400000
O	8.9095000000	9.9257400000	1.1087600000
H	9.6816700000	10.3617100000	1.4818900000
O	2.7231700000	0.5420800000	1.6718300000
O	3.4832800000	2.4631500000	-1.2498400000
O	0.0666900000	6.9430000000	0.3253200000
O	5.2710000000	6.6827600000	-2.1784100000
H	4.3885300000	7.0903100000	-2.3724600000
O	5.8814100000	4.1223800000	1.6872300000
H	6.6769700000	3.6965800000	2.1148800000
Mn	2.6327700000	15.3280100000	-0.8682000000
O	1.1821000000	14.9747500000	0.3934900000
O	3.8819400000	15.0734000000	0.5503800000
H	0.7841200000	14.0883200000	0.2174300000
H	0.4690500000	15.5448800000	0.0579500000
O	2.5012700000	17.1625200000	-0.3234300000
H	4.7415500000	15.4268400000	0.1972500000
O	4.1613600000	15.6160100000	-2.0967500000
O	2.6774200000	13.5999100000	-1.1293300000
H	3.2182700000	17.8222000000	-0.1982900000

H	2.6845200000	13.3889500000	-2.0714900000
H	2.4957400000	13.0557000000	-0.3016900000
H	2.3976200000	16.9094800000	0.6385800000
O	1.4684300000	15.4942900000	-2.4491000000
H	1.7425200000	16.3766100000	-2.7905100000
H	4.0474200000	14.1356000000	0.5120100000
H	5.1044500000	15.3767400000	-1.8951900000
H	4.0766200000	16.1545300000	-2.9179900000
H	1.9287000000	14.9699500000	-3.1758700000

ERGO-$[Fe(H_2O)_6]^{2+}$

104

C	12.02339875	8.09792337	17.20049159
C	13.29301262	7.54865481	17.22459679
C	14.47643848	8.43519184	17.30743529
C	15.82636868	7.91266422	17.05156442
C	16.91898813	8.80272089	17.00252874
C	18.27387611	8.31988887	17.07612691
C	19.35348539	9.22209598	17.06698072
C	11.79726258	9.50753416	17.04803266
C	12.81135765	10.45055577	16.84268188
C	14.19052585	9.94736409	17.09823790
C	15.35058754	10.74777505	16.59975491
C	16.71527306	10.22893018	16.89226767
C	17.79339716	11.11535217	16.87608184
C	19.11343818	10.60716419	16.96734486
C	11.33683109	12.50007430	16.50962430
C	12.63685293	11.89912979	16.64073049
C	13.74816817	12.80691642	16.61062864
C	15.10461137	12.23510039	16.44790489
C	16.25341562	13.14509329	16.55038953
C	17.61986347	12.61392184	16.97479923
C	18.75937114	13.34769188	16.30410246
C	11.17512809	13.87668248	16.75653244
C	12.28680898	14.70257486	16.89416217
C	13.61576371	14.24048810	16.64737849
C	14.75865787	15.13507695	16.41913628
C	16.06450303	14.49555350	16.33682924
C	17.24586278	15.29136921	15.80200777
C	18.58406812	14.59459848	15.83803576
C	11.11064722	17.95861251	16.59126993
C	12.22985651	17.09544183	16.64316613
C	13.51212972	17.43533718	16.13012062
C	14.70793249	16.59015835	16.25800032
C	15.96103355	17.33328973	16.31092056
C	17.23486452	16.68351733	16.38089184
C	18.37090891	17.36860860	16.81225806
C	11.18924170	19.17199747	15.91910860
C	12.41300732	19.58094581	15.32516207
C	13.62579885	18.79210509	15.58543212
C	14.89499054	19.42194957	15.62453489
C	15.99618130	18.77282178	16.26502333
C	17.15041229	19.48186435	16.73877723
C	18.31200972	18.75358677	17.10165448
O	13.59004593	6.21980552	17.19123078

H	15.98355431	6.82062953	17.00886353
O	18.50426452	6.98446711	17.18248454
C	12.29920973	20.74438382	14.47059329
O	15.10615477	20.71055462	15.18721272
C	17.13297809	20.95230788	16.94604851
H	11.14302183	7.42797033	17.26048467
H	10.74105281	9.81929049	17.15174656
O	20.61332289	8.67752486	17.16436451
O	20.21652148	11.43094836	16.99979232
O	10.20326525	11.81298787	16.20336605
H	10.15855061	14.29947532	16.84815785
C	20.10282151	12.64033454	16.14398966
H	19.41651430	15.12757023	15.33947589
H	10.17067348	17.61409027	17.05893366
H	10.29401299	19.80575602	15.77439849
H	19.32067510	16.82603206	16.97916166
C	19.39385328	19.41384043	17.90123966
H	12.75421017	5.71001750	17.05693400
H	19.48669548	6.86514534	17.24869265
O	11.42282650	21.65694691	14.57150525
O	13.21470884	20.80555484	13.40974101
H	12.91757151	21.57694850	12.86021714
H	14.54051556	20.83958213	14.36814210
O	16.22644946	21.58785922	17.56582791
O	18.18726219	21.59444214	16.39417051
H	18.27885820	22.47095681	16.85035522
H	21.25312421	9.43021025	17.21810251
H	20.90438064	11.53304552	14.75584803
H	10.47373635	10.92528506	15.85730836
O	20.19556339	12.22208212	14.80075859
O	21.11955037	13.49577792	16.56238577
H	21.97184260	13.12980319	16.22138584
O	19.21079875	20.41030462	18.60711861
O	20.58883920	18.76284735	17.80295103
H	21.20230627	19.25131803	18.41177876
O	14.40728599	9.40684753	18.41865640
O	15.20310178	11.31320975	15.27115697
O	12.00222714	15.96988675	17.39640127
O	17.02843659	15.42546212	14.35820645
H	16.15537761	15.87531514	14.24712449
O	17.75335129	12.98769370	18.39592187
H	17.28943654	12.27992953	18.91141786
Fe	13.94499534	24.14928251	16.18713956
O	12.98164626	24.60317181	17.99728404
O	15.90166981	24.01464466	16.80864635
O	14.22165308	26.37001345	15.75791899
H	14.82659197	26.87546319	16.35400685
H	16.11945214	23.06163526	17.20019137
O	13.57490681	22.18688784	17.17768935
O	12.39905218	23.98612996	14.84256154
O	15.36606174	24.18818943	14.21302365
H	15.22652307	25.06879040	13.78558449
H	11.95165432	23.03976068	14.68035467
H	14.44998419	21.71267119	17.33838849
H	13.65011548	27.06088919	15.34280676
H	16.35927299	24.03471647	15.92607362
H	11.64688697	24.62280898	14.88564917
H	13.03121774	21.51266587	16.69099521

H	12.74578502	25.39037608	18.54004187
H	12.76554606	23.79486348	18.52567179
H	15.49026241	23.55913331	13.46288951

ERGO-$[Cu(H_2O)_6]^{2+}$

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C	12.11275922	7.90846026	17.19738051
C	13.38622239	7.35991921	17.24947737
C	14.56231667	8.26279961	17.32934468
C	15.93118231	7.79588193	17.09508591
C	17.00015785	8.69013984	17.02014958
C	18.38201544	8.23506183	17.08586773
C	19.45251646	9.15675141	17.02868818
C	11.86575785	9.30955829	17.03054209
C	12.88251527	10.25010342	16.83800597
C	14.25469579	9.79147183	17.14475454
C	15.39389074	10.60545105	16.62793446
C	16.76374726	10.11714378	16.91094489
C	17.81741728	11.02179252	16.84695863
C	19.16006579	10.53940569	16.90407883
C	11.36646801	12.21891643	16.55593347
C	12.69356404	11.68017832	16.66706059
C	13.78005693	12.61688531	16.68296749
C	15.13385249	12.08741795	16.47425160
C	16.26026944	13.01381194	16.56897472
C	17.62725939	12.51146988	16.95296120
C	18.75951552	13.27443239	16.31060161
C	11.18565458	13.56552336	16.80987922
C	13.62481760	14.03838494	16.70922792
C	14.74727013	14.94906744	16.49937342
C	16.06366112	14.34412047	16.36146649
C	17.22675464	15.17209946	15.86694772
C	18.56750221	14.52336965	15.88169194
C	11.01552040	17.66693982	16.55838757
C	12.14147473	16.84176095	16.67401791
C	13.47330221	17.22758235	16.38260664
C	14.67357418	16.38419523	16.46323578
C	15.92998606	17.10629266	16.57466560
C	17.21774581	16.50150614	16.54608898
C	18.36240671	17.19909499	16.89951244
C	11.12183540	18.91122889	15.95924775
C	12.38308968	19.37926513	15.55275324
C	13.56686127	18.60418489	15.90041002
C	14.86251395	19.14693488	15.93720457
C	15.95984674	18.52742291	16.59227359
C	17.14042614	19.27122351	16.92819678
C	18.32598396	18.58655644	17.19347162
O	13.70031955	6.03013328	17.30424409
H	16.10548623	6.70696930	17.10735180
O	18.56693262	6.90168714	17.22803858
C	12.23963212	20.66392315	14.84835400
O	15.25623559	20.27218724	15.26195973
C	17.03347679	20.74766664	17.20840022
H	11.24942902	7.21231550	17.22072430
H	10.79740369	9.61127606	17.08632650
O	20.72799955	8.63379984	17.08123870

O	20.25925023	11.36827629	16.90531293
O	10.19453045	11.57103943	16.26938114
H	10.18211790	14.02729786	16.83211639
C	20.09678229	12.61370864	16.05863437
H	19.38931900	15.07390676	15.39256219
H	10.04472281	17.21881981	16.83930600
H	10.19344904	19.46070833	15.72881322
H	19.33755314	16.69125162	16.95465683
C	19.49478734	19.33350718	17.76253870
H	12.87272790	5.51059870	17.13665576
H	19.53677157	6.70924739	17.29707641
O	11.46242274	21.56213801	15.19834388
O	13.02938229	20.74715355	13.65080445
H	12.73917042	21.60432635	13.23331103
H	14.92834200	20.18033440	14.33479920
O	16.34956457	21.19589136	18.13721280
O	17.64832804	21.47486243	16.20720764
H	17.66519834	22.42425647	16.50045078
H	21.36610525	9.39581185	17.11168728
H	20.71122929	11.43020675	14.63219811
H	10.41385192	10.82861679	15.65968526
O	20.12217830	12.21929731	14.70797409
O	21.14379512	13.43529276	16.43826747
H	21.91801527	13.15808299	15.89310091
O	19.41248699	20.48884691	18.18308227
O	20.62323878	18.55379603	17.75896374
H	21.34116847	19.11156567	18.16236177
O	14.47518396	9.23754258	18.44852384
O	15.22218631	11.15270516	15.28964930
O	11.74742777	15.65786753	17.21107423
O	17.01682843	15.40370789	14.41947969
H	16.11633898	15.80558374	14.33822015
O	17.59085503	12.83839377	18.40313211
H	18.39105653	12.41469979	18.79986783
Cu	14.30343536	24.24161398	15.83036746
O	12.73139541	23.67356486	17.24106117
O	15.76541989	23.81126952	17.41655155
H	12.45251156	22.78340196	16.89694099
H	12.11851028	24.27679128	16.75142994
O	14.15963131	26.08477722	16.46162068
H	16.52521987	24.17624875	16.91449551
O	15.95947569	24.33522342	14.50891116
O	14.43745214	22.35211844	15.58168284
H	14.99743818	26.60381491	16.46325950
H	14.37035024	21.92266554	14.69578527
H	14.21581934	21.64989989	16.23677756
H	14.10181927	25.66100084	17.36128937
O	13.09301963	24.24409937	14.05134222
H	13.41126605	25.16022728	13.86143092
H	15.81753602	22.80890454	17.30627469
H	16.89985552	24.11545278	14.69584877
H	15.92070503	24.90733454	13.71284289
H	13.63728620	23.66699731	13.46649446

ERGO-$[H_2PO_4]^-$

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C	12.07227457	8.28426574	17.09508163
C	13.34078182	7.75751656	17.18488471
C	14.52083390	8.64587348	17.29027844
C	15.87052504	8.13897491	17.05933154
C	16.94457748	9.04862645	16.97515744
C	18.31360717	8.60516744	17.01404176
C	19.36904792	9.51936095	16.91497296
C	11.82358990	9.69246727	16.90541148
C	12.83035142	10.64173690	16.72982231
C	14.21015809	10.16165023	17.04312226
C	15.36240185	10.95923303	16.53542053
C	16.72689417	10.46972293	16.82288503
C	17.79250448	11.38106134	16.72941849
C	19.11282806	10.90477107	16.78483175
C	11.33962393	12.67533752	16.37207317
C	12.63714055	12.09253572	16.51093945
C	13.74301554	13.01358121	16.50067559
C	15.10036011	12.45905911	16.32745932
C	16.22674689	13.38654437	16.41839679
C	17.59598674	12.87439460	16.82825321
C	18.72883619	13.63880923	16.17306162
C	11.15406329	14.04112727	16.64045455
C	12.25352618	14.87366830	16.83783525
C	13.59029744	14.44342341	16.58582259
C	14.71841156	15.36987021	16.40429725
C	16.02320129	14.74812083	16.27876248
C	17.19938278	15.58555751	15.78907290
C	18.54036143	14.90413563	15.76645117
C	10.99465639	17.98542738	16.35058775
C	12.14865753	17.23775869	16.59185132
C	13.45465082	17.67498880	16.22436175
C	14.65345378	16.83141566	16.33578845
C	15.90062464	17.56843135	16.42435467
C	17.19077326	16.93723415	16.45330322
C	18.32510859	17.64235277	16.82717770
C	11.08597394	19.27727749	15.80464062
C	12.33581259	19.83784177	15.52280570
C	13.53281631	19.05853614	15.75809974
C	14.84611373	19.67170068	15.73477326
C	15.93349488	18.99591153	16.39733665
C	17.08196214	19.73420493	16.82428347
C	18.26168564	19.03920999	17.12972124
O	13.62626774	6.40970148	17.17330278
H	16.04999843	7.05195588	17.05060862
O	18.58192599	7.26306942	17.17856619
C	12.27402993	21.29320996	15.07397358
O	15.14894073	20.79228154	15.14278500
C	16.87095763	21.22649092	16.99652846
H	11.20335502	7.59437876	17.11271270
H	10.76023342	9.99129494	16.95849564
O	20.66151128	9.01439346	16.97910298
O	20.22634855	11.75030383	16.78532739
O	10.21239694	11.96394872	16.02880326
H	10.13244780	14.45443393	16.72790285

C	20.07436248	12.96067578	15.96453018	
H	19.36035610	15.47383623	15.28927098	
H	10.02431499	17.55912704	16.66724874	
H	10.18975094	19.90869609	15.65821005	
H	19.30307162	17.12984142	16.88576234	
C	19.43309443	19.74936873	17.70499888	
H	12.78050858	5.94014837	16.97993437	
H	19.56881459	7.19939166	17.21758595	
O	11.38206289	22.04480179	15.52850000	
O	13.17170764	21.69626744	14.18313164	
H	13.82829858	23.18176340	14.24320131	
H	14.22361000	21.19339037	14.57092742	
O	15.99485250	21.63533877	17.78175035	
O	17.59989968	22.00552486	16.18085965	
H	17.14887278	22.91563342	16.20641211	
H	21.25109499	9.80931880	16.98499996	
H	20.65635852	11.71038896	14.60482256	
H	10.53821102	11.09958483	15.67291012	
O	20.19411493	12.58393313	14.59626418	
O	21.11567019	13.81668306	16.34683172	
H	21.87426019	13.56520148	15.76602295	
O	19.41469010	20.86209067	18.23374748	
O	20.59339890	19.00321185	17.60819334	
H	21.26666608	19.58204391	18.04819017	
O	14.38679649	9.64970900	18.38312687	
O	15.19380211	11.48504306	15.19223045	
O	11.96559806	16.11573899	17.38872568	
O	16.97691591	15.82404232	14.35296661	
H	16.14322006	16.35537636	14.30576243	
O	17.63507429	13.21552993	18.27376168	
H	18.09893625	12.45763280	18.70738771	
P	14.39098669	24.61650760	15.82867466	1
O	13.20936299	23.83341027	16.64189653	
O	15.73476335	23.99094327	16.57990378	
O	14.41401816	26.11012613	15.86976525	
H	15.51476563	23.16960942	17.14120644	
H	12.49776948	23.41348252	16.06620794	
O	14.37361756	24.03957198	14.31662571	

ERGO-As(OH)$_3$

92

C	12.10320340	8.16042135	17.09583290
C	13.37253580	7.63933020	17.18735935
C	14.54366690	8.53624226	17.28387197
C	15.89372588	8.02976366	17.04663905
C	16.96906287	8.93200329	16.96585760
C	18.33361420	8.47691755	17.01969807
C	19.39578721	9.38416725	16.92459482
C	11.85416790	9.56752753	16.90276738
C	12.85549186	10.52041835	16.71450638
C	14.23533471	10.05099928	17.02123070
C	15.38535484	10.84386463	16.51261881
C	16.74777127	10.35153075	16.80721991
C	17.81357948	11.25363550	16.72461217
C	19.13517782	10.76661266	16.78954580

C	11.36473306	12.54819138	16.36191366
C	12.66410979	11.97113210	16.49970655
C	13.76765158	12.89423682	16.49650734
C	15.12738756	12.34579984	16.32458270
C	16.25592537	13.26839752	16.43267821
C	17.62706643	12.75015540	16.82878033
C	18.75213983	13.50694146	16.15106898
C	11.18080843	13.91689468	16.62299658
C	12.27775195	14.75187604	16.82613703
C	13.61611070	14.32444017	16.58376843
C	14.74613863	15.25052073	16.40377296
C	16.05188941	14.62921851	16.29486394
C	17.22229265	15.45903488	15.77543322
C	18.56736746	14.77615693	15.75326592
C	11.02123929	17.89553097	16.41498670
C	12.17329398	17.12197068	16.59562605
C	13.47684744	17.53938953	16.18744137
C	14.68094427	16.71432342	16.30487984
C	15.92698048	17.47153210	16.36781390
C	17.21151166	16.83329250	16.39752472
C	18.34901983	17.53140953	16.77342963
C	11.10353824	19.16191809	15.81329024
C	12.34754625	19.67803039	15.43334684
C	13.56132463	18.92425300	15.71718321
C	14.82201606	19.57958414	15.79994800
C	15.94948886	18.90993366	16.35968652
C	17.11851010	19.63702467	16.79143032
C	18.29165158	18.92709116	17.07233845
O	13.68828633	6.30381877	17.18187149
H	16.06277653	6.94050177	16.99478630
O	18.58228399	7.14379149	17.19138509
C	12.26903963	21.01830698	14.80003064
O	15.02543167	20.86485860	15.41154414
C	16.96189733	21.12299913	17.06935733
H	11.23789590	7.46811588	17.11523067
H	10.79053271	9.86125116	16.95753363
O	20.67621165	8.86623150	16.99757914
O	20.23756845	11.61036910	16.79067303
O	10.24164656	11.82841821	16.03231662
H	10.15794859	14.32425546	16.70991423
C	20.08664160	12.81183677	15.92706959
H	19.38615974	15.34080686	15.26824208
H	10.06124336	17.48399848	16.77421284
H	10.19769242	19.77315925	15.64233793
H	19.32228766	17.01525991	16.84969601
C	19.47709232	19.62520310	17.65231435
H	12.86407867	5.79654442	16.99068379
H	19.56673781	7.04887277	17.22370891
O	11.50701918	21.93819549	15.11661644
O	13.12497960	21.16989842	13.69069803
H	12.98315253	22.10984845	13.39654165
H	14.43452636	21.05801478	14.62377153
O	16.20094156	21.49515863	17.98280451
O	17.59639880	21.94162457	16.21470035
H	17.18801726	22.86442711	16.39434965
H	21.28410430	9.64624919	17.00052276
H	20.76812655	11.60597513	14.56818728
H	10.56156151	10.95967112	15.68265425

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O     20.15319268      12.38090758      14.58210726
O     21.13920104      13.66098139      16.27602644
H     21.91734890      13.36539583      15.74386929
O     19.47787373      20.75658666      18.13835824
O     20.60372349      18.83112538      17.61105650
H     21.29958352      19.38688037      18.04571406
O     14.43126441       9.54855191      18.35938015
O     15.23071911      11.38702583      15.17963509
O     11.98881720      15.99663286      17.37887790
O     16.98244292      15.63640607      14.33519711
H     16.12496265      16.12524848      14.26488858
O     17.67590739      13.08822459      18.26879687
H     18.37001023      12.50128071      18.66062068
As    14.47950179      24.01365897      15.74715880
O     13.23828496      23.58384400      17.01127947
O     15.90515302      23.91801080      16.90814731
O     14.40292280      25.82331079      15.77276645
H     14.61066725      26.06890555      16.71120800
H     15.75339886      23.12356843      17.53164286
H      12.51038739      23.11244791     16.52328483
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(d) Force-field optimized geometries of oxidation mechanism steps for the cluster-cutout model of Figure 8 in the manuscript

Initial state

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C         -0.68487         8.88926        -0.18791
C          0.45417         8.13186        -0.00823
C          1.75541         8.55299        -0.39652
C          2.95253         7.74098        -0.27199
C          4.18453         8.49764        -0.20906
C         -0.60162        10.15663        -0.77534
C          0.62792        10.67839        -1.12160
C          1.83569         9.93853        -0.82446
C          3.07105        10.59968        -0.71189
C          4.20242         9.92929        -0.20956
C          0.56704        12.01273        -1.76324
O          3.19949        11.92668        -0.99920
H         -1.62887         8.49219         0.16706
H         -1.49746        10.74356        -0.94728
O         -0.16729        12.92746        -1.43887
O          1.36562        12.13187        -2.87923
H          1.20382        13.03108        -3.22770
H          2.86943        12.08916        -1.90491
O          0.28285         7.00927         0.75472
As         2.72239        15.14463        -0.78007
O          1.50043        14.69819         0.46497
O          4.14151        15.00383         0.34666
O          2.66056        16.93431        -0.73496
H          3.97295        14.28940         1.00560
H          0.78616        14.20087         0.02304
H          2.92701         6.71008        -0.23171
H          5.08067         7.98797        -0.16223
H          4.99109        10.18867        -0.88456
```

H	1.00930	6.44193	0.92317
H	2.88190	16.98777	0.08488

Step 1

29

C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	3.07105	10.59968	-0.71189
C	4.20242	9.92929	-0.20956
C	0.56704	12.01273	-1.76324
O	3.19949	11.92668	-0.99920
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
O	-0.16729	12.92746	-1.43887
O	1.36562	12.13187	-2.87923
H	1.20382	13.03108	-3.22770
H	2.86943	12.08916	-1.90491
O	0.28285	7.00927	0.75472
As	2.52196	15.02689	-1.18335
O	1.66614	16.57922	-0.86637
O	3.65251	15.18856	0.23050
O	3.73186	15.51476	-2.41093
H	4.28639	16.22035	-2.02982
H	0.73181	16.46798	-1.12614
H	2.92701	6.71008	-0.23171
H	5.08067	7.98797	-0.16223
H	4.99109	10.18867	-0.88456
H	1.00930	6.44193	0.92317

Step 2

29

C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	3.07105	10.59968	-0.71189
C	4.20242	9.92929	-0.20956
C	0.55748	12.05492	-1.75667
O	3.29135	11.87573	-1.12709
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
O	-0.44229	12.87916	-1.21452

O	0.37393	11.94397	-3.13544
H	1.15283	12.41177	-3.53048
H	2.77014	12.52508	-1.59426
O	0.28285	7.00927	0.75472
As	0.32922	13.90283	0.11049
O	-0.91229	15.15642	0.63808
O	1.12216	14.30226	1.72349
O	0.57670	14.95259	-1.38082
H	1.55027	15.13317	-1.39636
H	-1.72425	14.63112	0.85209
H	2.92701	6.71008	-0.23170
H	5.08067	7.98797	-0.16223
H	4.33822	10.24711	0.80309
H	1.00930	6.44193	0.92317

Step 3

29

C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	2.81221	10.70774	-0.44878
C	4.20242	9.92929	-0.20956
C	1.14071	12.03053	-1.45127
O	2.51936	11.92023	-1.04603
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
O	0.34204	12.91014	-0.68748
O	1.01475	12.27844	-2.82376
H	1.55821	13.08534	-3.01329
O	0.28285	7.00927	0.75472
As	1.11844	14.51430	-0.22489
O	-0.25000	15.56230	0.41781
O	2.29237	14.29226	1.16465
O	1.86662	15.37397	-1.66516
H	2.84037	15.24353	-1.55252
H	-0.93537	14.91370	0.72426
H	2.55219	10.91848	0.59046
H	2.92701	6.71008	-0.23170
H	5.08067	7.98797	-0.16223
H	4.58185	10.24220	0.74072
H	1.00930	6.44193	0.92317

Step 4

29

C	-0.6848700000	8.8892600000	-0.1879100000
C	0.4541700000	8.1318600000	-0.0082300000
C	1.7554100000	8.5529900000	-0.3965200000
C	2.9525300000	7.7409800000	-0.2719900000
C	4.1845300000	8.4976400000	-0.2090600000
C	-0.6016200000	10.1566300000	-0.7753400000
C	0.6279200000	10.6783900000	-1.1216000000
C	1.8356900000	9.9385300000	-0.8244600000
C	2.8122100000	10.7077400000	-0.4487800000
C	4.2024200000	9.9292900000	-0.2095600000
C	1.1407100000	12.0305300000	-1.4512700000
O	2.5193600000	11.9202300000	-1.0460300000
H	-1.6288700000	8.4921900000	0.1670600000
H	-1.4974600000	10.7435600000	-0.9472800000
O	0.3420400000	12.9101400000	-0.6874800000
O	1.0147500000	12.2784400000	-2.8237600000
H	1.5582100000	13.0853400000	-3.0132900000
O	0.2828500000	7.0092700000	0.7547200000
As	1.1184400000	14.5143000000	-0.2248900000
O	-0.2500000000	15.5623000000	0.4178100000
O	2.2923700000	14.2922600000	1.1646500000
O	1.8666200000	15.3739700000	-1.6651600000
H	-0.9353700000	14.9137000000	0.7242600000
H	2.5521900000	10.9184800000	0.5904600000
H	2.9270100000	6.7100800000	-0.2317000000
H	5.0806700000	7.9879700000	-0.1622300000
H	4.5818500000	10.2422000000	0.7407200000
H	-0.2745200000	13.0496000000	-1.4232200000
H	1.0093000000	6.4419300000	0.9231700000

Final step

29

C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	2.99235	10.60783	-0.70561
C	4.20242	9.92929	-0.20956
C	0.60763	12.03393	-1.79800
O	-0.43858	12.66283	-1.87978
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
O	-0.83935	15.07075	0.05360
O	1.70148	12.54223	-2.40464
H	1.67942	13.39377	-2.86394
O	0.28285	7.00927	0.75472
As	0.81015	15.88288	0.07326

O	0.69242	17.52345	0.89593
O	1.99871	14.81031	0.97757
O	1.37932	16.09870	-1.66166
H	2.30382	15.74401	-1.66738
H	-0.17435	17.50225	1.37442
H	3.07215	11.66553	-0.87410
H	2.92701	6.71008	-0.23170
H	5.08067	7.98797	-0.16223
H	4.36445	10.25149	0.79783
H	1.00930	6.44193	0.92317

(e) Force-field optimized geometries of oxidation mechanism steps for the full-sheet model

Initial state

92

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024
C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975
C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811
C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793
C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	3.07105	10.59968	-0.71189
C	4.20242	9.92929	-0.20956

C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	0.56704	12.01273	-1.76324
O	3.19949	11.92668	-0.99920
C	5.23819	12.12107	0.51335
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495
C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	-0.16729	12.92746	-1.43887
O	1.36562	12.13187	-2.87923
H	1.20382	13.03108	-3.22770
H	2.86943	12.08916	-1.90491
O	4.57238	12.50871	1.45824
O	5.83312	12.91015	-0.37822
H	5.53004	13.83466	-0.18694
H	9.54686	0.69774	0.45476
H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600
H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	2.72239	15.14463	-0.78007
O	1.50043	14.69819	0.46497
O	4.14151	15.00383	0.34666
O	2.66056	16.93431	-0.73496
H	2.87140	17.22456	0.17164
H	3.97295	14.28940	1.00560
H	0.78616	14.20087	0.02304

Step 1

91

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024
C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975
C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811
C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793
C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	3.07105	10.59968	-0.71189
C	4.20242	9.92929	-0.20956
C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	0.56704	12.01273	-1.76324
O	3.19949	11.92668	-0.99920
C	5.23819	12.12107	0.51335
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495
C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901

H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	-0.16729	12.92746	-1.43887
O	1.36562	12.13187	-2.87923
H	1.20382	13.03108	-3.22770
H	2.86943	12.08916	-1.90491
O	4.57238	12.50871	1.45824
O	5.83312	12.91015	-0.37822
H	5.53004	13.83466	-0.18694
H	9.54686	0.69774	0.45476
H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600
H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	2.52196	15.02689	-1.18335
O	1.66614	16.57922	-0.86637
O	3.65251	15.18856	0.23050
O	3.73186	15.51476	-2.41093
H	3.19401	15.65117	0.97131
H	0.73181	16.46798	-1.12614

Step 2

91

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024
C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975
C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811

C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793
C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	3.07105	10.59968	-0.71189
C	4.20242	9.92929	-0.20956
C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	0.55748	12.05492	-1.75667
O	3.29135	11.87573	-1.12709
C	5.26971	12.12412	0.51094
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495
C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	-0.44229	12.87916	-1.21452
O	0.37393	11.94397	-3.13544
H	1.15283	12.41177	-3.53048
H	2.77014	12.52508	-1.59426
O	4.28202	12.57057	1.08083
O	6.20858	12.98670	0.06257
H	6.13317	13.94022	0.21069
H	9.54686	0.69774	0.45476
H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600

H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	0.32922	13.90283	0.11049
O	-0.91229	15.15642	0.63808
O	1.12216	14.30226	1.72349
O	0.57670	14.95259	-1.38082
H	1.55027	15.13317	-1.39636
H	-1.72425	14.63112	0.85209

Step 3

91

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024
C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975
C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811
C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793

C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	2.81221	10.70774	-0.44878
C	4.20242	9.92929	-0.20956
C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	1.14071	12.03053	-1.45127
O	2.51936	11.92023	-1.04603
C	5.26971	12.12412	0.51094
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495
C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	0.34204	12.91014	-0.68748
O	1.01475	12.27844	-2.82376
H	1.55821	13.08534	-3.01329
O	4.28202	12.57057	1.08083
O	6.20858	12.98670	0.06257
H	6.13317	13.94022	0.21069
H	9.54686	0.69774	0.45476
H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600
H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	1.11844	14.51430	-0.22489
O	-0.25000	15.56230	0.41781
O	2.29237	14.29226	1.16465
O	1.86662	15.37397	-1.66516
H	2.84037	15.24353	-1.55252
H	-0.93537	14.91370	0.72426
H	2.55219	10.91848	0.59046

Step 4

91

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024
C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975
C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811
C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793
C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	2.81221	10.70774	-0.44878
C	4.20242	9.92929	-0.20956
C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	1.14071	12.03053	-1.45127
O	2.51936	11.92023	-1.04603
C	5.26971	12.12412	0.51094
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495

C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	0.34204	12.91014	-0.68748
O	1.01475	12.27844	-2.82376
H	1.55821	13.08534	-3.01329
O	4.28202	12.57057	1.08083
O	6.20858	12.98670	0.06257
H	6.13317	13.94022	0.21069
H	9.54686	0.69774	0.45476
H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600
H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	1.11844	14.51430	-0.22489
O	-0.25000	15.56230	0.41781
O	2.29237	14.29226	1.16465
O	1.86662	15.37397	-1.66516
H	2.84037	15.24353	-1.55252
H	-0.93537	14.91370	0.72426
H	2.89937	10.74265	0.67654

Final state

91

C	0.42181	-0.78880	0.52151
C	1.67326	-1.30753	0.60469
C	2.84237	-0.41873	0.70213
C	4.18779	-0.90998	0.45707
C	5.25137	-0.01320	0.38024
C	6.61301	-0.46210	0.41230
C	7.65914	0.44231	0.31361
C	0.17100	0.61393	0.33201
C	1.15505	1.55870	0.15366
C	2.53292	1.09828	0.45258
C	3.67162	1.88888	-0.04975

C	5.02857	1.40116	0.23837
C	6.08348	2.29934	0.15375
C	7.39157	1.81449	0.19747
C	-0.33111	3.57930	-0.17970
C	0.95710	3.00958	-0.06811
C	2.05376	3.92844	-0.08335
C	3.40847	3.38603	-0.25143
C	4.52463	4.30708	-0.14472
C	5.88709	3.79334	0.27026
C	7.01167	4.55560	-0.38994
C	-0.51439	4.93848	0.05752
C	0.57091	5.76831	0.23459
C	1.89996	5.34964	-0.00406
C	3.02402	6.27427	-0.18022
C	4.31757	5.65794	-0.28405
C	5.48707	6.49162	-0.78378
C	6.82839	5.81173	-0.77942
C	-0.68487	8.88926	-0.18791
C	0.45417	8.13186	-0.00823
C	1.75541	8.55299	-0.39652
C	2.95253	7.74098	-0.27199
C	4.18453	8.49764	-0.20906
C	5.46226	7.86199	-0.16793
C	6.58694	8.54967	0.20821
C	-0.60162	10.15663	-0.77534
C	0.62792	10.67839	-1.12160
C	1.83569	9.93853	-0.82446
C	2.99235	10.60783	-0.70561
C	4.20242	9.92929	-0.20956
C	5.37344	10.64229	0.22429
C	6.53144	9.93947	0.49947
O	1.97658	-2.63439	0.58758
H	4.35419	-1.97651	0.38508
O	6.86068	-1.78580	0.55918
C	0.60763	12.03393	-1.79800
O	-0.43858	12.66283	-1.87978
C	5.26971	12.12412	0.51094
H	-0.43636	-1.45809	0.53167
H	-0.87275	0.89514	0.36497
O	8.93946	-0.05446	0.35480
O	8.48289	2.65199	0.17784
O	-1.46667	2.86844	-0.44648
H	-1.51950	5.33486	0.14495
C	8.33863	3.86300	-0.62976
H	7.63139	6.36936	-1.24901
H	-1.62887	8.49219	0.16706
H	-1.49746	10.74356	-0.94728
H	7.53997	8.04228	0.28633
C	7.71462	10.63645	1.07329
H	1.16913	-3.14234	0.40605
H	7.82599	-1.90535	0.60772
O	-0.83935	15.07075	0.05360
O	1.70148	12.54223	-2.40464
H	1.67942	13.39377	-2.86394
O	4.28202	12.57057	1.08083
O	6.20858	12.98670	0.06257
H	6.13317	13.94022	0.21069
H	9.54686	0.69774	0.45476

H	8.97201	2.68250	-2.02524
H	-1.22007	2.06958	-0.93950
O	8.40727	3.47326	-1.97362
O	9.38293	4.69012	-0.26600
H	10.15687	4.44216	-0.79875
O	7.71219	11.75839	1.53452
O	8.83477	9.86896	1.05058
H	9.53103	10.41131	1.46525
O	2.73738	0.58054	1.75737
O	3.51337	2.42077	-1.36403
O	0.28285	7.00927	0.75472
O	5.28809	6.66939	-2.22104
H	4.41716	7.08378	-2.34189
O	5.89867	4.10572	1.70258
H	6.67456	3.66277	2.09136
As	0.81015	15.88288	0.07326
O	0.69242	17.52345	0.89593
O	1.99871	14.81031	0.97757
O	1.37932	16.09870	-1.66166
H	2.30382	15.74401	-1.66738
H	-0.17435	17.50225	1.37442
H	3.07215	11.66553	-0.87410

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