SUPPLEMENTARY INFORMATION

Arsenic immobilization as crystalline scorodite by gas-diffusion electrocrystallization.

G. Pozo[1,3], D. Van Houtven[1,3], J. Fransaer[2,3] and X. Dominguez-Benetton[1,3]

[1]*Separation and Conversion Technologies, VITO-Flemish Institute for Technological Research, Boeretang 200, 2400, Mol, Belgium.*

[2]*Department of Materials Engineering, Surface and Interface Engineered Materials, Katholieke Universiteit Leuven, Kasteelpark Arenberg 44 - box 2450, 3001 Leuven, Belgium.*

[3] *SIM vzw, Technologiepark 935,BE-9052 Zwijnaarde, Belgium.*

* *Correspondence should be addressed to:*

Xochitl Dominguez-Benetton, Separation and Conversion Technologies, VITO, Flemish Institute for Technological Research, Boeretang 200, 2400, Mol, Belgium

Phone: +3214336926; E-mail: xoch@vito.be

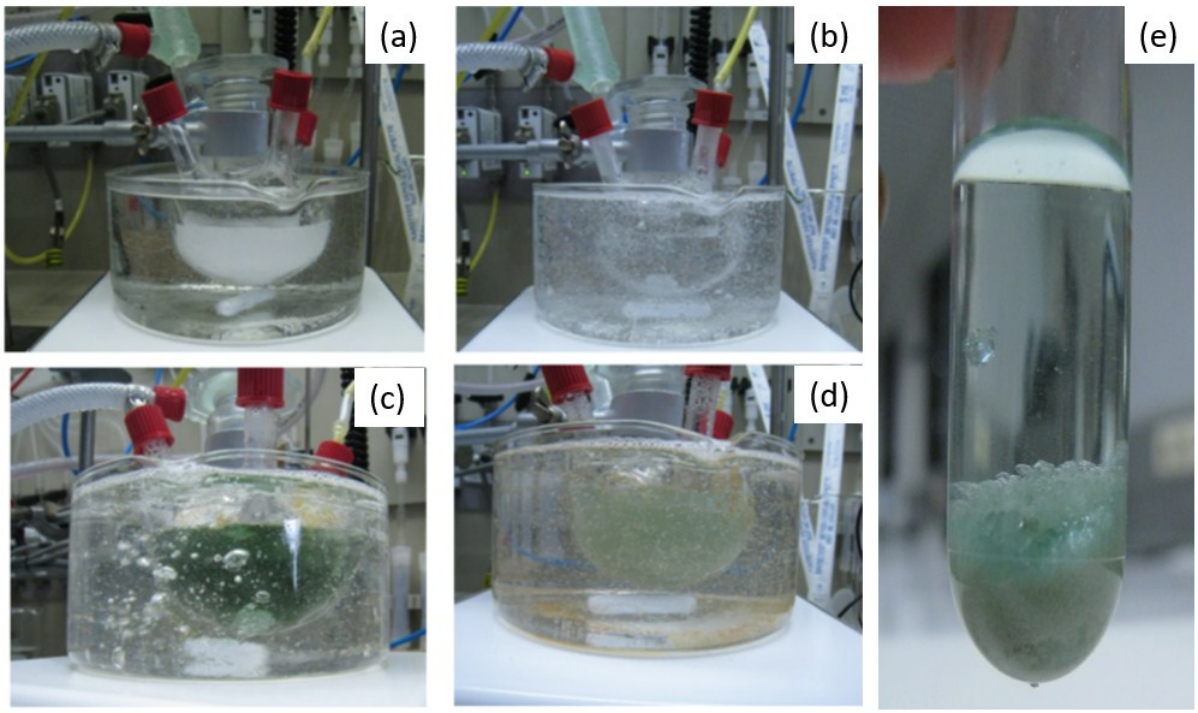

Figure ESI1. Experimental sequence for the chemical precipitation of scorodite. (a) Insoluble 0.22 M As^{5+} at room temperature (~18 °C), (b) setup at 50 °C showing As^{5+} dissolved. (c) Solution turning into green color after the addition of 1.25 M Fe^{2+}, d) after 18 hours at 95 °C with a constant flow of O_2 and (e) green-gray precipitates formed by chemical precipitation.

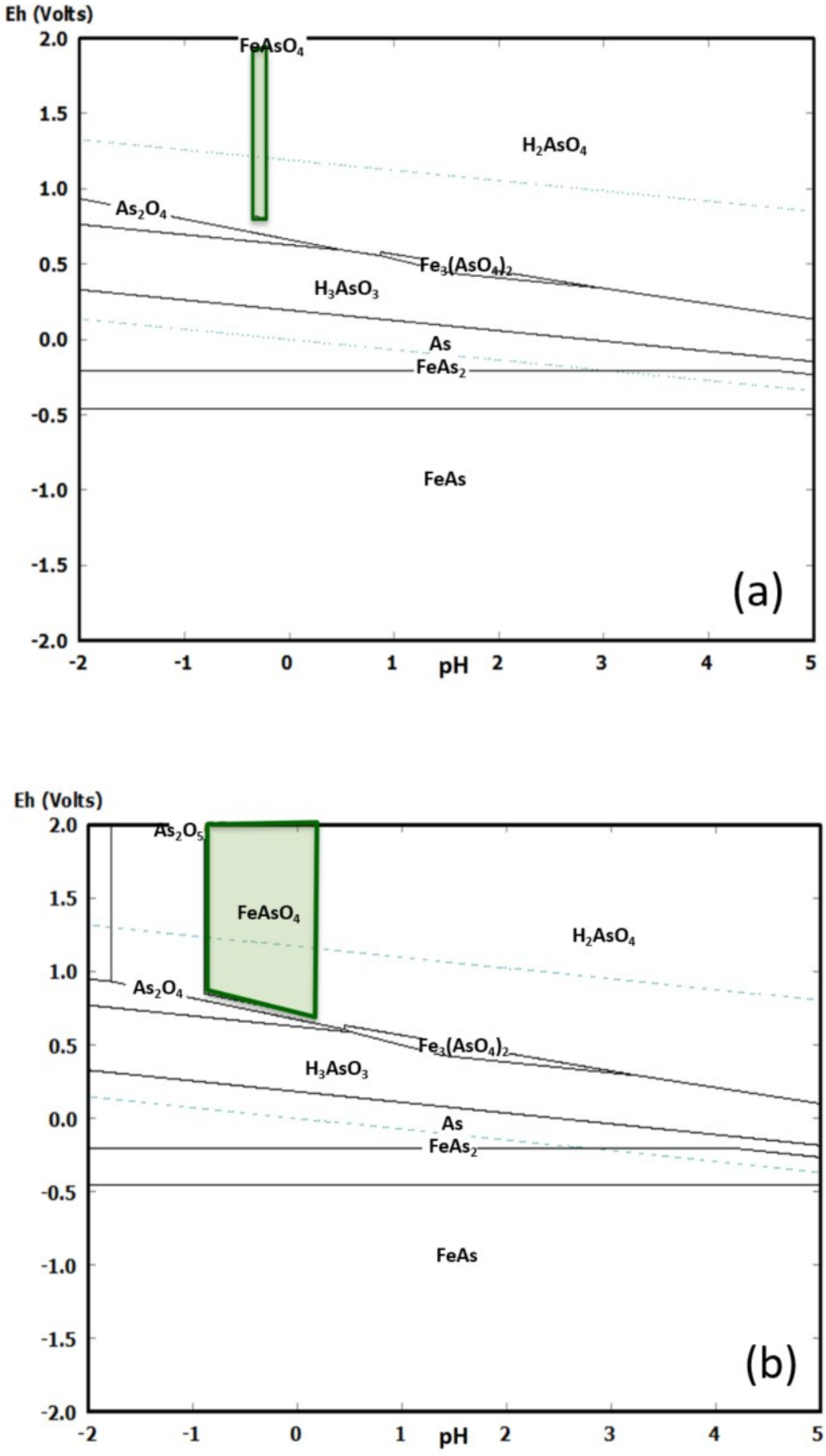

Figure ESI2. E-pH diagrams for As-Fe-H_2O system (Σ As = 0.04 M and Σ Fe = 0.27 M) at a) 70 °C and b) 95 °C using HSC-Chemistry 9.0 software.

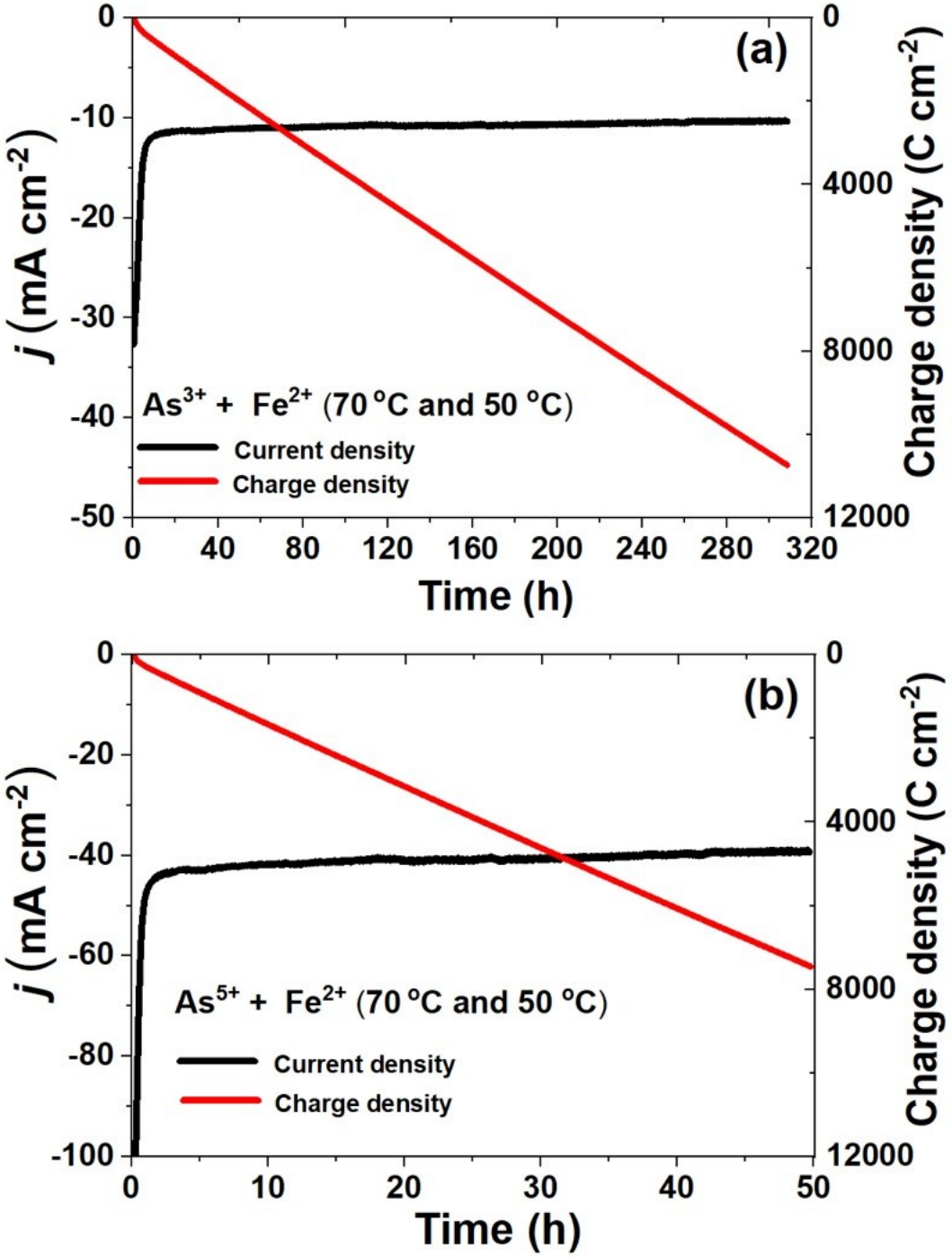

Figure ESI3. Chronoamperometry test at -0.15 $V_{SHE.}$ a) As^{3+} and Fe^{2+} as precursors at 50 °C and 70 °C and b) As^{5+} and Fe^{2+} as precursors at 50 °C and 70 °C.

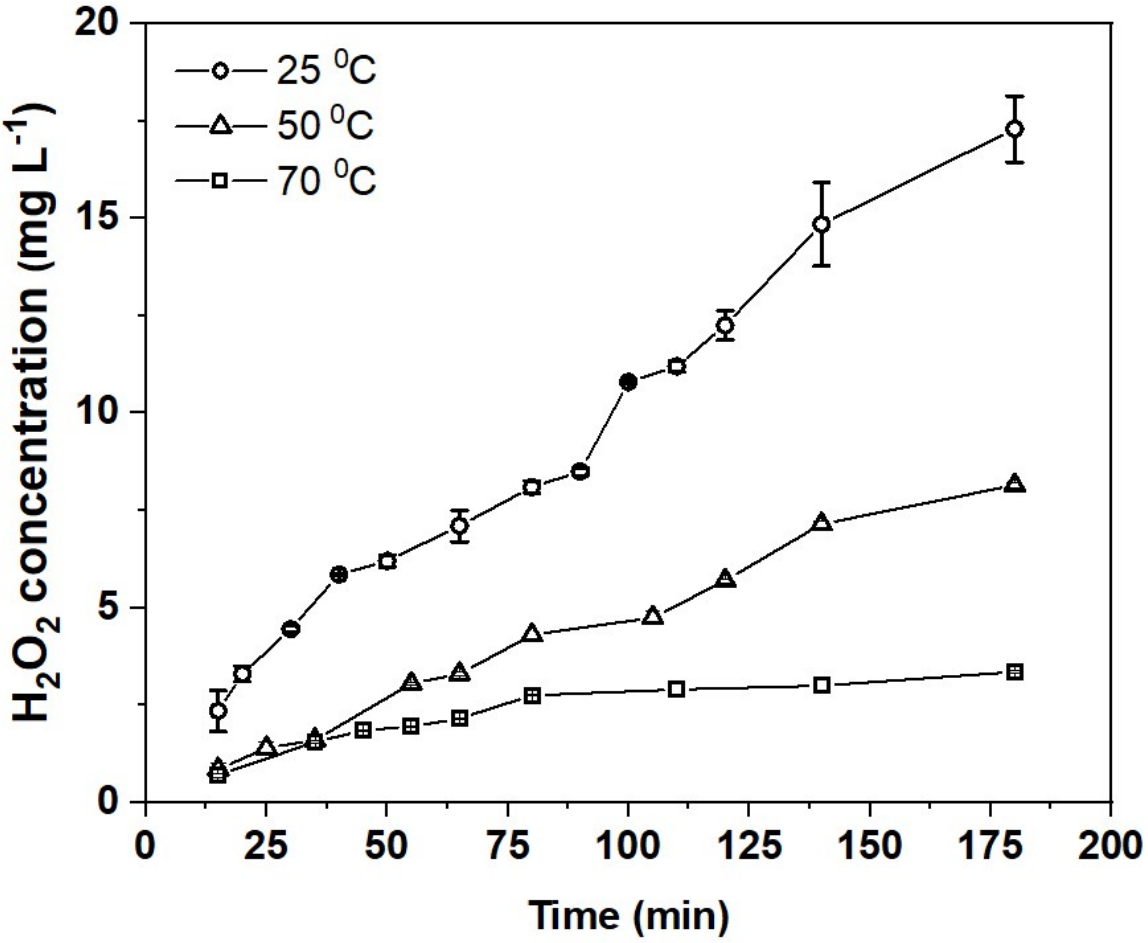

Figure ESI4. Effect of temperature on the production of H_2O_2 after the electrochemical reduction of oxygen at -0.15V_{SHE} using 0.3 M Na_2SO_4 at pH 2.2 (pH adjusted with H_2SO_4) at 25 °C, 50 °C and 70 °C.

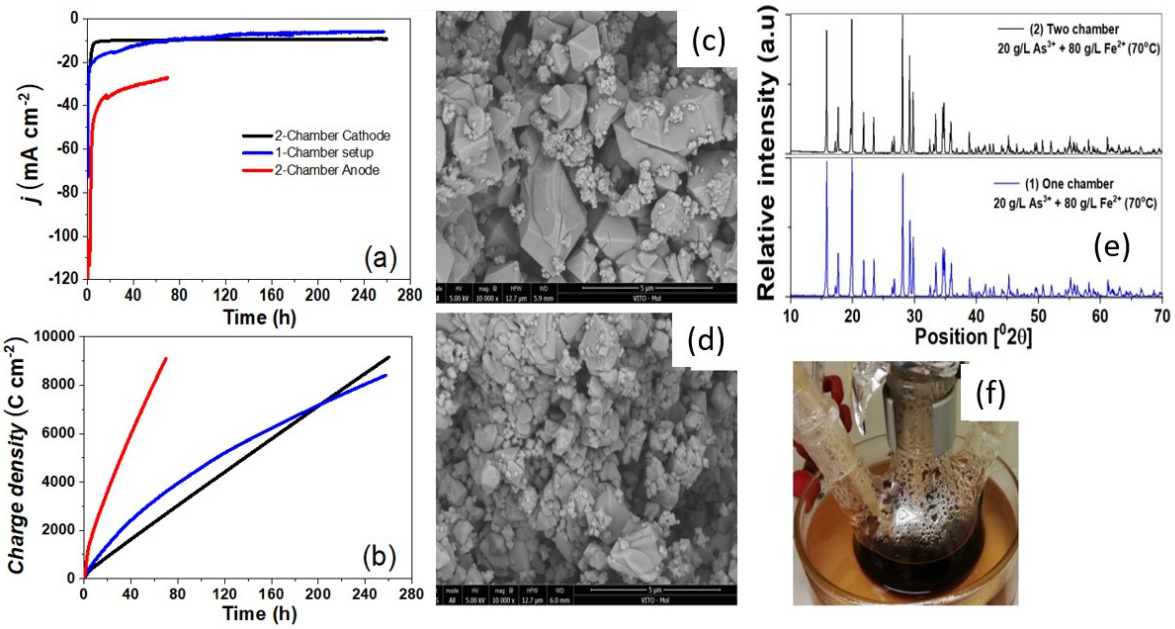

Figure ESI5. Comparison of the three GDEx operation modes and product characterization. Current densities (a) and charge consumption (b) under the three different strategies. SEM image of scorodite made with two-chamber GDEx (c) and one-chamber GDEx (d). XRD pattern (black) of scorodite precipitate in the cathode side (GDE side) in a two-chamber setup and with one chamber reactor (e). Anolyte solution after the two-chamber operation, where the As^{3+} and Fe^{2+} precursor were fed in the anode side without scorodite precipitation (f).

Table ESI1. Summary of the GDEx operational conditions using different precursors and synthesis temperatures.

	Precursor			
	As^{3+} , Fe^{2+}	**As^{3+} , Fe^{2+}**	**As^{5+} , Fe^{2+}**	**As^{5+} , Fe^{2+}**
Temperature (°C)	50	70	50	70
pH	0.0	0.0	0.0	0.0
Conductivity (mS cm^{-1})	122	122	153	153
Total charge consumed (C)	107,350	96,713	77,131	74,540
Processing time (h)	308	304	53	50
Current density (mA cm^{-2})	10 ± 2	9 ± 3	39 ± 7	41 ± 1
Charge density (C cm^{-2})	10,735	9,671	7,713	7,454

Table ESI2. Summary of the GDEx operational conditions at different feeding strategies.

Experiment	**Two-chamber setup, wherein As^{3+} and Fe^{2+} were supplied to the cathode chamber (side of the GDE)**	**One-chamber setup without a Nafion membrane**	**Two-chamber GDEx by feeding the precursor in the anode side without contact with the GDE**
Precursor	As^{3+}, Fe^{2+}	As^{3+}, Fe^{2+}	As^{3+}, Fe^{2+}
Temperature (^{0}C)	70	70	70
Total charge consumed (C)	91,550	84,050	90,920
Processing time (h)	260	257	70
Current density (mA cm^{-2})	9 ± 3	9 ± 4	34 ± 8
Charge density (C cm^{-2})	9,155	8,405	9,092

Table ESI3. Crystallite size and lattice parameters obtained from XRD data through Rietveld refinement of scorodite synthesized under different conditions.

Precursor	Temperature (°C)	Reactor setup	Crystallite size (nm)	Lattice parameters (nm)		
				a	b	c
As^{3+} , Fe^{2+}	50	Two-chamber	71 ± 2	0.89	1.03	1
As^{3+} , Fe^{2+}	70	Two-chamber	108 ± 0	0.89	1.03	1
As^{5+} , Fe^{2+}	50	Two-chamber	70 ± 6	0.90	1.03	1
As^{5+} , Fe^{2+}	70	Two-chamber	66 ± 4	0.90	1.03	1
As^{3+} , Fe^{2+}	70	One-chamber	79 ± 3	0.90	1.03	1