SUPPLEMENTARY MATERIAL

Manganese-Cobalt Geomimetic Materials for Supercapacitor Electrode

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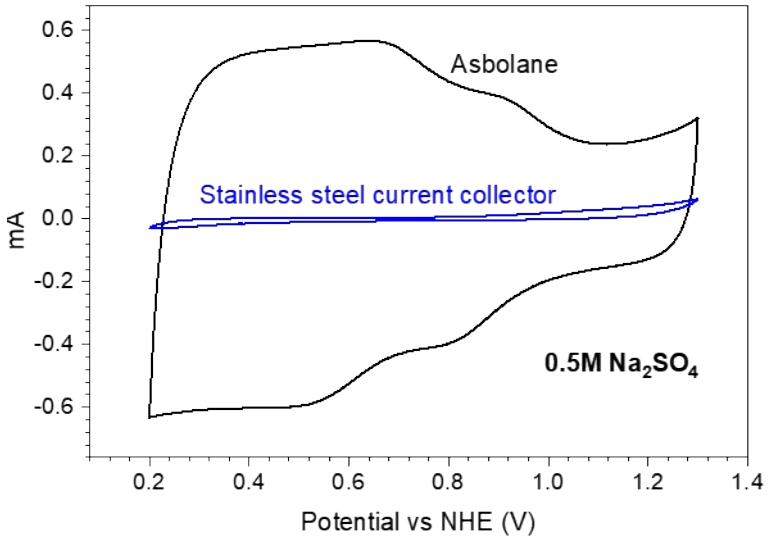


Figure S1 : Comparison of the CV curves of bare stainless-steel current collector with the one of Asbolane in 0.5M Na_2SO_4 at 5 mV/s. The capacity of bare current collector, which is directly proportional to the area of the voltammogram is negligible (lower than 2%) compared to this of asbolane material.

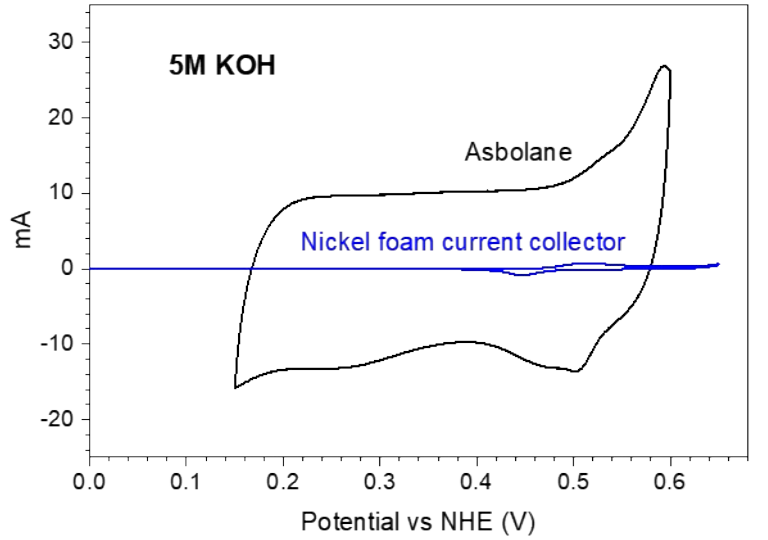


Figure S2 : Comparison of the CV curves of bare stainless-steel current collector with the one of Asbolane in 5M KOH at 10 mV/s. The capacity of bare current collector, which is directly proportional to the area of the voltammogram is negligible compared to this of asbolane material.

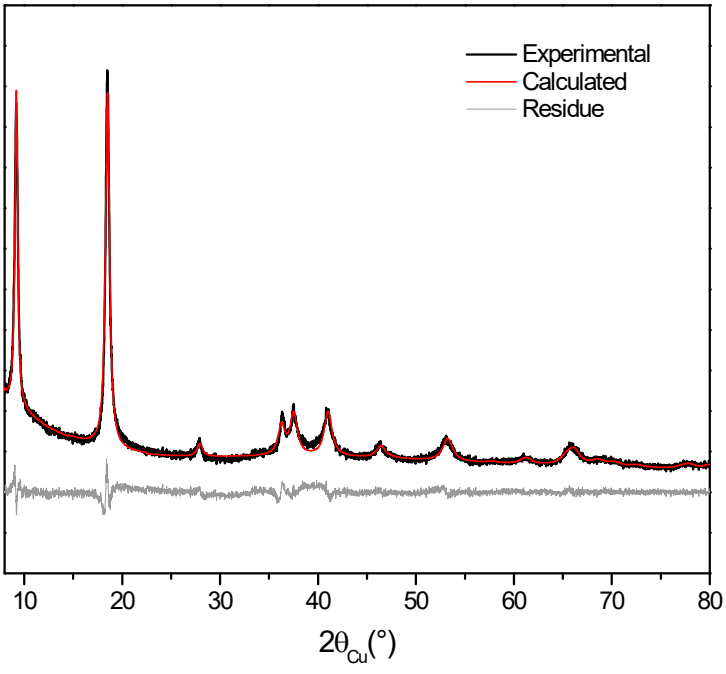


Figure S3 : Results of the Le Bail refinement for asbolane. The black curve corresponds to the experimental pattern and the red one to the calculated profile. The residue (grey curve) is also shown.

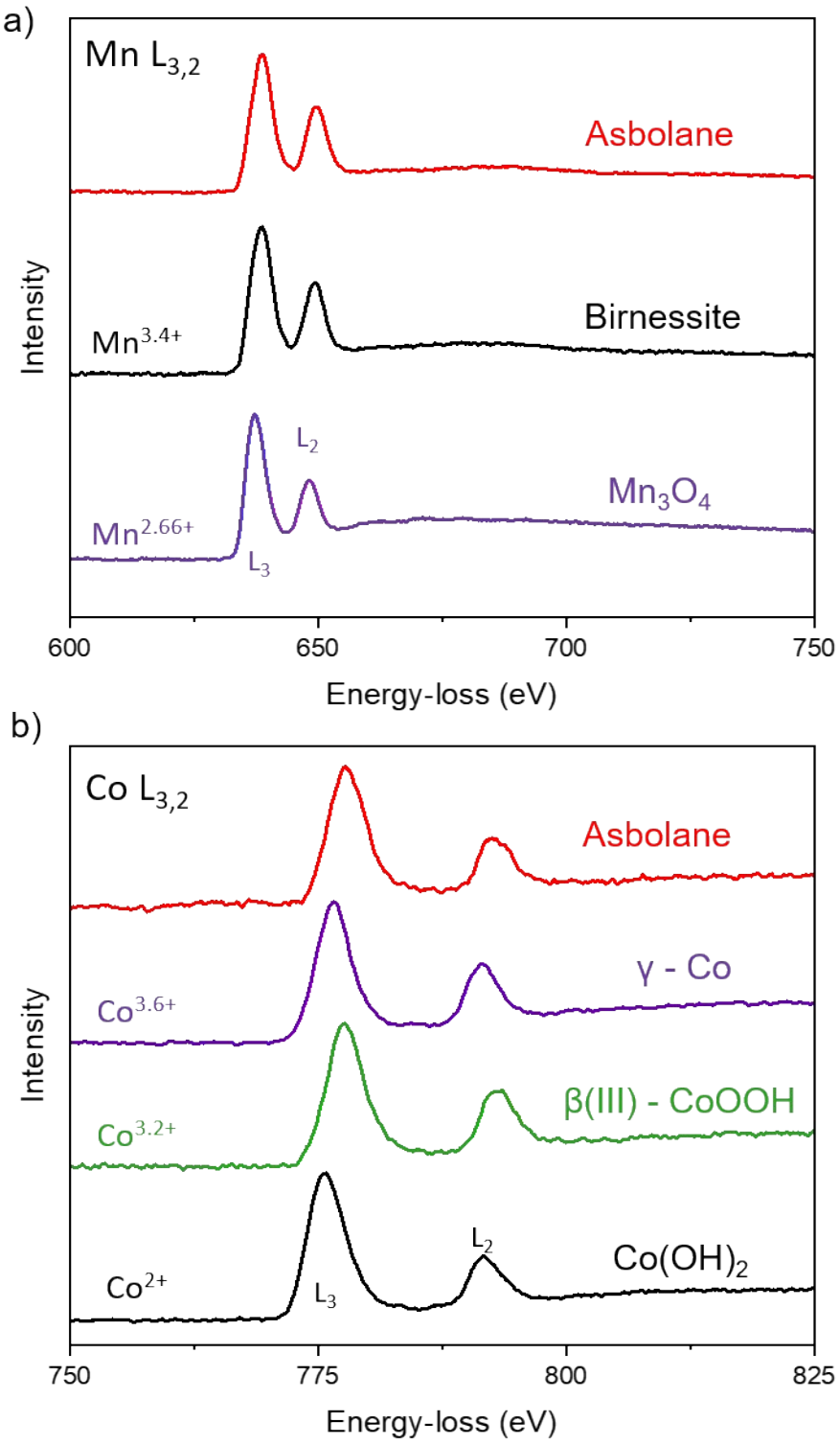


Figure S4 : Normalized EELS spectra for the references (Mn_3O_4, Birnessite, $Co(OH)_2$, β(III)-CoOOH, γ – Co) and Asbolane phases for a) Mn $L_{3,2}$, b) Co $L_{3,2}$ edges and c) Table listing the oxidation degree of each materials and the calculated values of the intensity ratio L3/L2.

	Oxidation degree	L_3/L_2 (Mn) Intensity	L_3/L_2 (Co) Intensity
Mn_3O_4	Mn: +2.66	2.78	/
Birnessite	Mn: +3.4	2.26	/
$Co(OH)_2$	Co: +2	/	2.72
β(III) - CoOOH	Co: +3.2	/	2.31
γ- Co	Co: +3.6	/	2.24
Asbolane	Presence of Co^{2+}, Co^{3+}, Mn^{3+}, Mn^{4+}	2.21	2.53

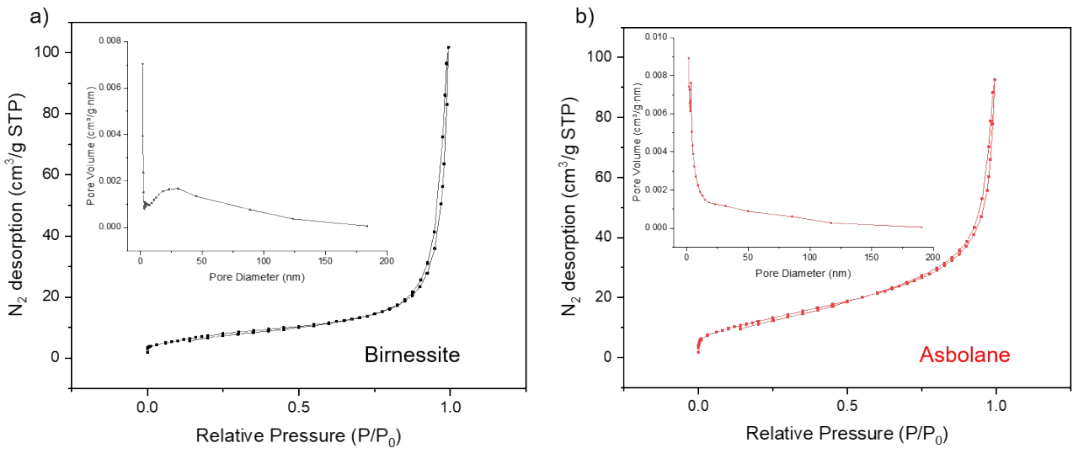
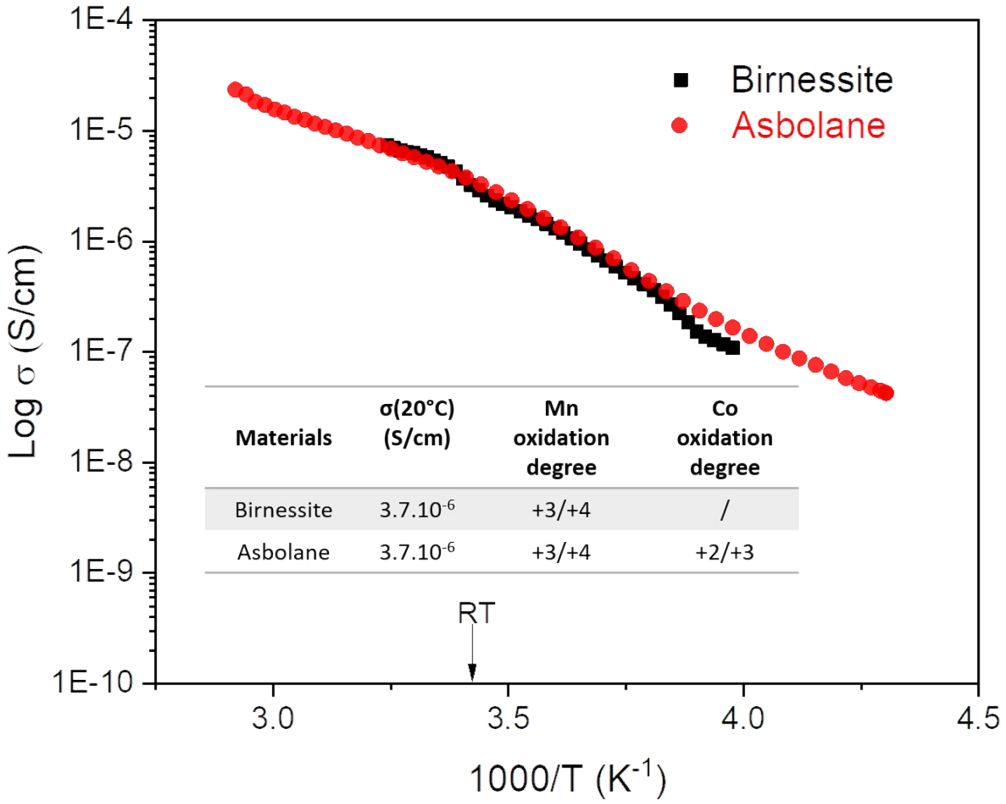


Figure S5 : N_2 adsorption/desorption isotherms obtained as well as the curves representing the pore size distribution as a function of their volume for a) Birnessite and b) Asbolane phases.



Materials	σ(20°C) (S/cm)	Mn oxidation degree	Co oxidation degree
Birnessite	$3.7.10^{-6}$	+3/+4	/
Asbolane	$3.7.10^{-6}$	+3/+4	+2/+3

Figure S6 : Thermal variation of electronic conductivity and table summarizing the electronic conductivity values at 20°C as well as the oxidation degree of cobalt and manganese for Birnessite and Asbolane materials.

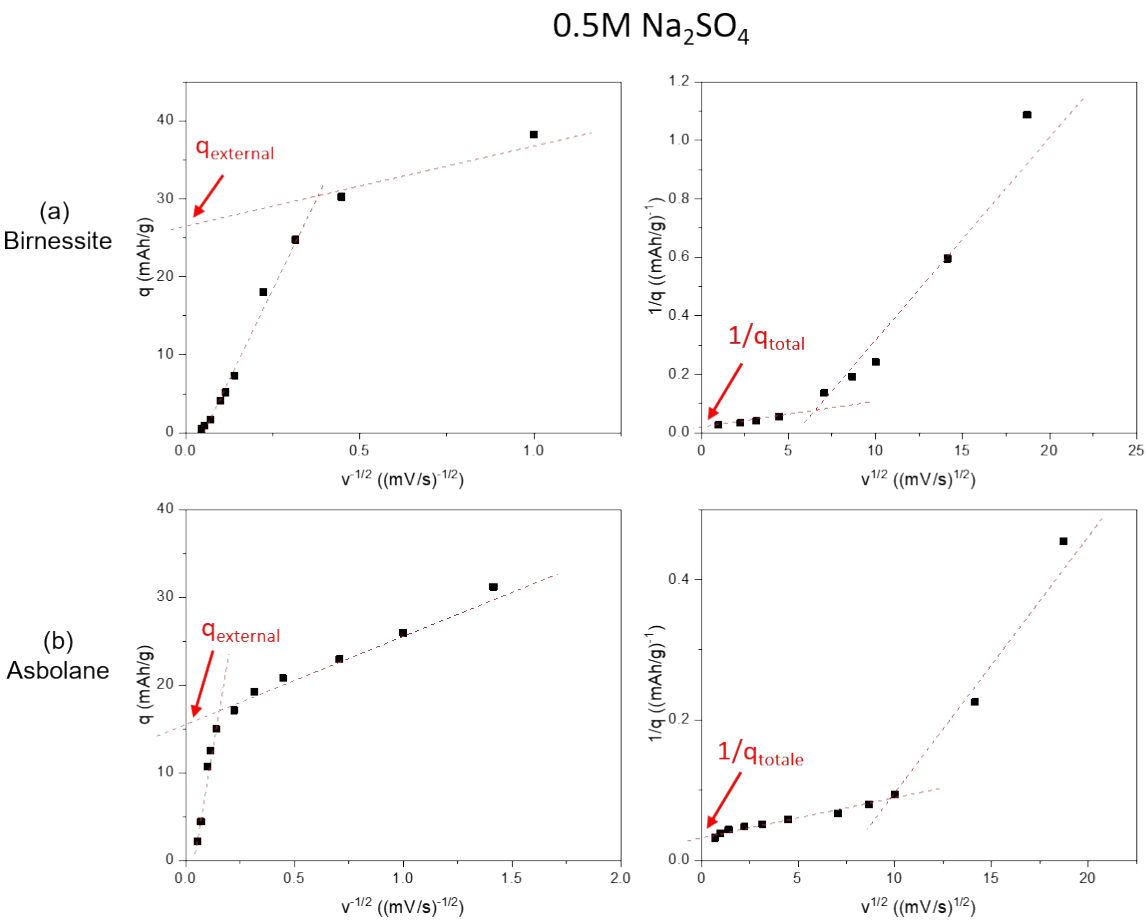


Figure S7 : Extrapolations at $v \rightarrow +\infty$ of $q = f(v^{-1/2})$ and at $v \rightarrow 0$ of $1/q = f(v^{1/2})$ for a) Birnessite and b) Asbolane materials for scan rates ranging from 0.5 to 350 mV/s in 0.5M Na_2SO_4.

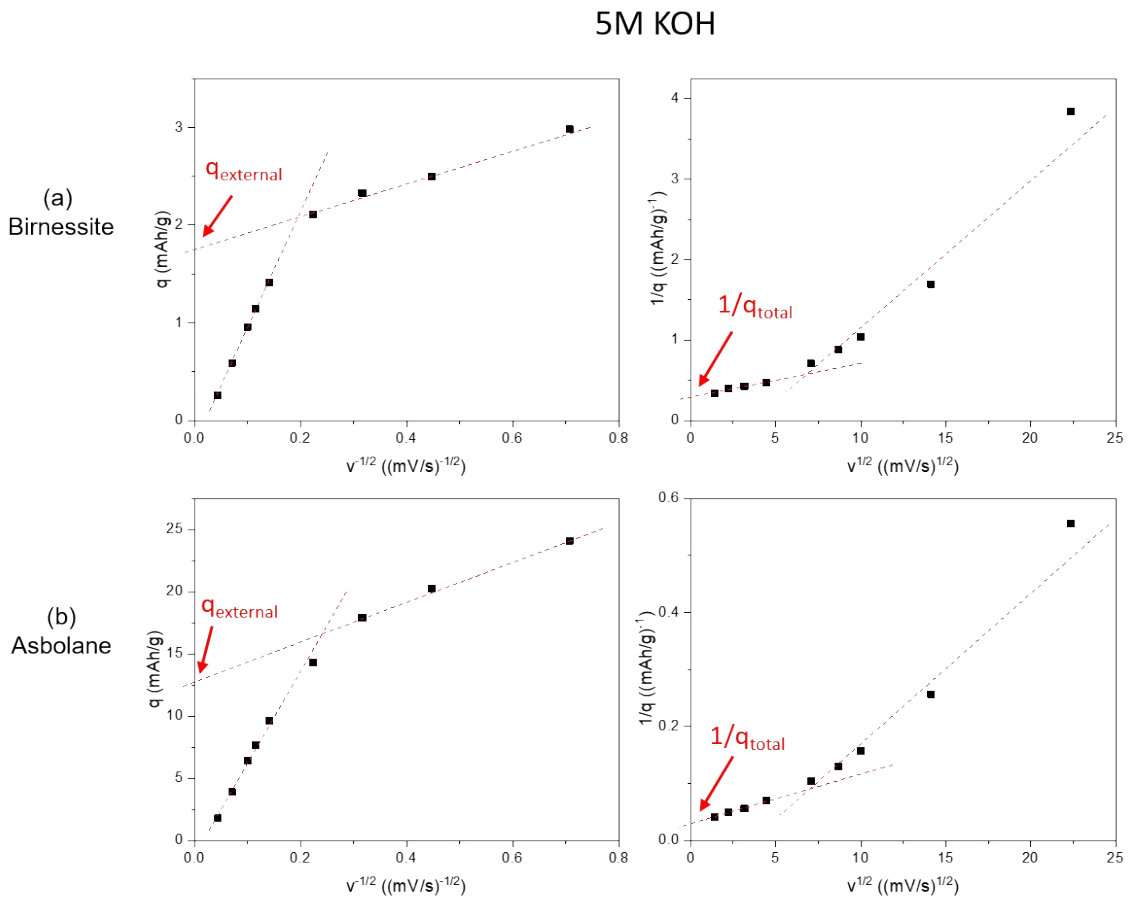


Figure S8 : Extrapolations at $v \rightarrow +\infty$ of $q = f(v^{-1/2})$ and at $v \rightarrow 0$ of $1/q = f(v^{1/2})$ for a) Birnessite and b) Asbolane materials for scan rates ranging from 2 to 500 mV/s in 5M KOH.

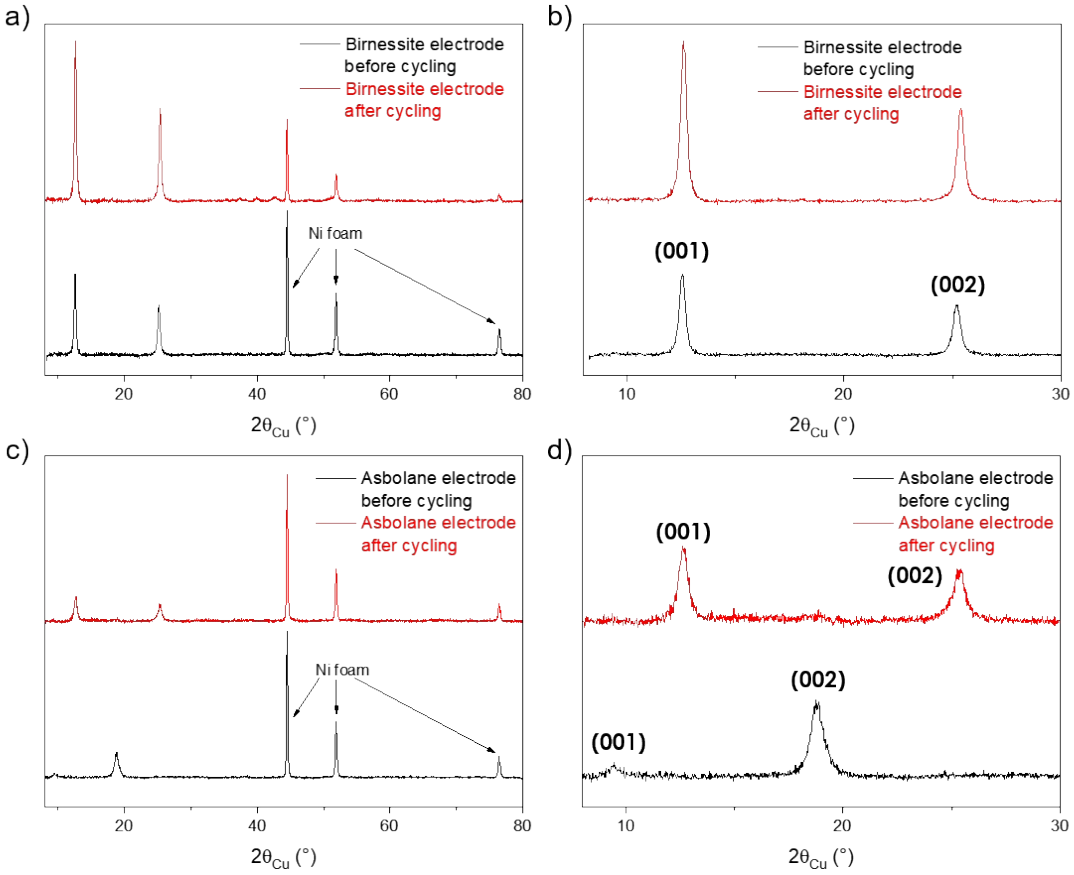


Figure S9 : X-ray diffractograms obtained before and after 3000 galvanostatic charge/discharge cycles at 1A/g in 5M KOH for a-b) Birnessite and c-d) Asbolane.

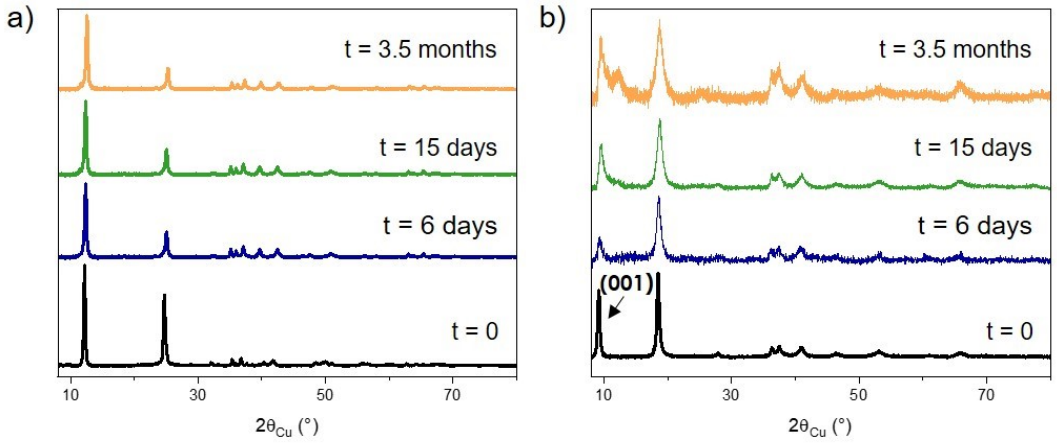


Figure S10 : X-ray diffractograms obtained before and after aging during 6 days, 15 days and 3.5 months in 5M KOH at room temperature for a) Birnessite and b) Asbolane materials.

Table S1 : Parameters used for simulating the X-ray diffraction profiles with the Diffax software.

	2 layers model	1 layer model
Wavelength	λ_{Cu} = 1.5406 Å	λ_{Cu} = 1.5406 Å
Point Group Symmetry	-3M	-3M
Unit Cell Parameters	a = b = 2.84 Å c = 9.6 Å α = β= 90° γ= 120°	a = b = 2.84 Å c = 9.6 Å α = β= 90° γ= 120°
Instrumental Function	Pseudo-Voigt (Gaussian and Lorentzian contribution)	Pseudo-Voigt (Gaussian and Lorentzian contribution)
Atom Positions Notation used: Considered atom: x, y, z and occupancy	MnO_2 layer = layer 1 O : 2/3 1/3 0.6 1 Mn : 0 0 0.5 1 O : 1/3 2/3 0.4 1 CoO_2 layer = layer 2 O : 2/3 1/3 0.6 1 Co : 0 0 0.5 0.55 O : 1/3 2/3 0.4 1	Layer 1 : O : 0 0 0.7 1 Co : 1/3 2/3 0.6 0.55 O : 2/3 1/3 0.5 1 Mn : 0 0 0.4 1 O : 1/3 2/3 0.3 1
Translation Vectors Notation used: Transition probability, $\vec{x}, \vec{y}, \vec{z}$	Layer 1 to Layer 1 : 0 0 0 0 Layer 1 to Layer 2 : 1 0 0 z_1 Layer 2 to Layer 1 : 1 0 0 z_2 Layer 2 to Layer 2 : 0 0 0 0	Layer 1 to Layer 1 : 1 0 0 1

Table S2: Binding Energies (eV) and atomic percentages (at. %) extracted from XPS data for Birnessite and Asbolane.

	Birnessite		Asbolane	
Orbitals	B.E. (eV)	at. %	B.E. (eV)	at. %
C 1s	285.0 286.5 288.6	13.7 2.6 1.9 18.2	285.0 286.7 288.8	17.7 2.2 2.3 22.2
O 1s	530.0 531.6 533.0 534.6	39.5 7.7 3.1 2.8 53.2	530.4 531.6 532.8	27.7 20.6 6.0 54.3
N 1s			398.7 400.3 403.6 407.2	0.1 0.2 0.1 0.7 1.8
Ni 2p				
Mn 2p (Mn^{+4}) Mn 2p (Mn^{+3})	642.6 641.1	9.3 11.2 20.5	642.7 641.1	10.7 3.4 14.1
Co 2p (Co^{+3}) Co 2p (Co^{+2})			780.4 781.0	2.4 6.0 8.4
Na 1s		8.1		

Table S3 : q_{total}, $q_{external}$ and $q_{internal}$ values (in mAh/g and F/g) determined by Trasatti method in 5M KOH and 0.5M Na_2SO_4 for Birnessite and Asbolane.

	0.5M Na_2SO_4			**5M KOH**		
	q_{total}	$q_{external}$	$q_{internal}$	q_{total}	$q_{external}$	$q_{internal}$
Birnessite	47.5 mAh/g (~156 F/g)	47.5 mAh/g (~156 F/g)	47.5 mAh/g (~156 F/g)	3.4 mAh/g (~27 F/g)	1.7 mAh/g (~13.5 F/g)	1.7 mAh/g (~13.5 F/g)
Asbolane	31.3 mAh/g (~102 F/g)	15.5 mAh/g (~51 F/g)	15.8 mAh/g (~51 F/g)	34.5 mAh/g (~276 F/g)	12.7 mAh/g (~27 F/g)	21.8 mAh/g (~13.5 F/g)