

## Supporting Information

### **Pathways Towards High Performance Na-O<sub>2</sub> Batteries: Tailoring Graphene Aerogel Cathode Porosity & Nanostructure**

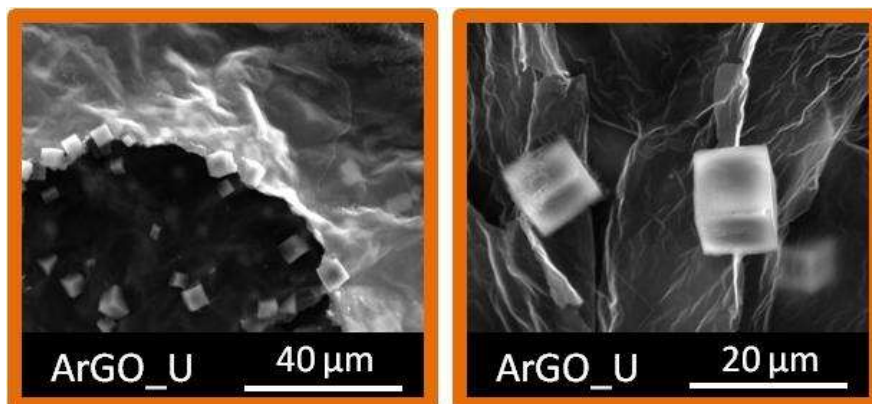
M. Enterría<sup>1</sup>, C. Botas<sup>1</sup>, J.L. Gómez-Urbano<sup>1</sup>, B. Acebedo<sup>1</sup>, Juan Miguel López del Amo<sup>1</sup>,  
D. Carriazo<sup>1,2</sup>, T. Rojo<sup>1,3</sup>, N. Ortiz-Vitoriano<sup>1,2,\*</sup>

<sup>1</sup>*CIC EnergiGUNE, Álava Technology Park. C/ Albert Einstein 48, 01510 Miñano Spain*

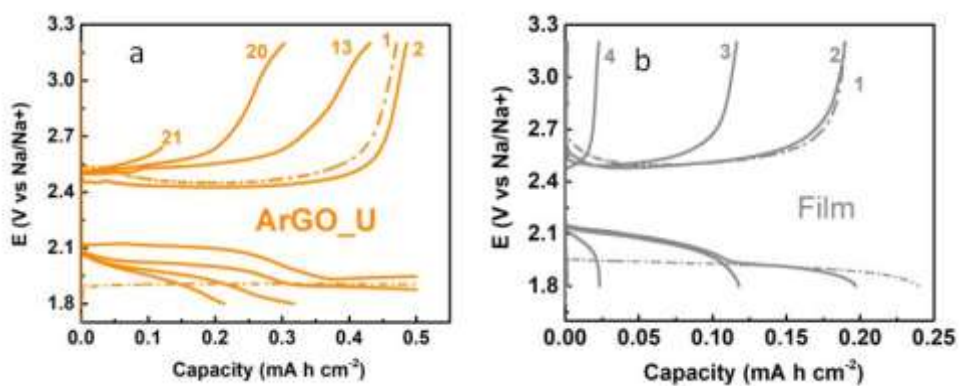
<sup>2</sup>*IKERBASQUE, Basque Foundation for Science, 48013 Bilbao, Spain*

<sup>3</sup>*Departamento de Química Inorgánica, Universidad del País Vasco UPV/EHU, P.O. Box  
664, 48080, Bilbao, Spain*

\*Correspondence to [nortiz@cicenergigune.com](mailto:nortiz@cicenergigune.com)



**Fig. S1.** SEM images of galvanostatically discharged ArGO\_U cathode at 100 mAh g<sup>-1</sup> to 1.8 V showing the O<sub>2</sub>/electrode interface.



**Fig. S2.** a) Cyclability test for ArGO\_U and (b) Film electrodes at 100 mA/g and 0.5 mAh cm<sup>-2</sup> using 0.1M NaClO<sub>2</sub> in DME as electrolyte.

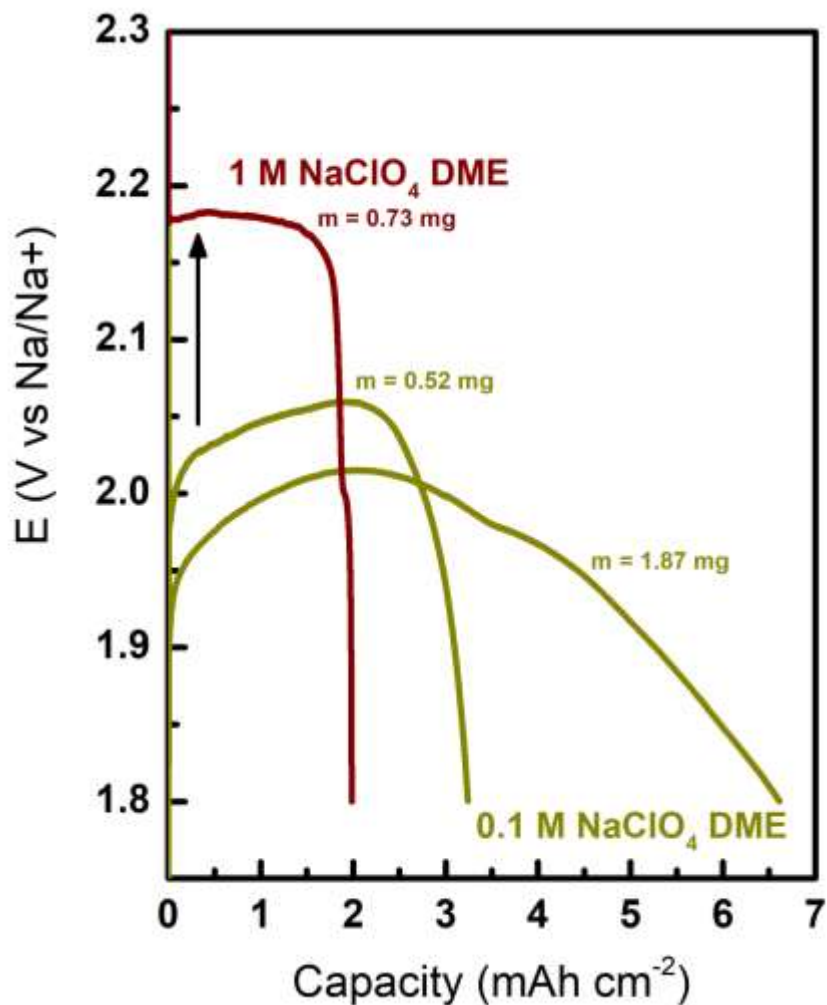
**Table S1.** Electrochemical Properties of Na-air/O<sub>2</sub> batteries produced by this work and from literatures.

Air-cathode [loading (mg) and area (cm <sup>2</sup> )]	Electrolyte/ Environment	Discharge properties			Cycling performance			
		Capacity	Discharge product	Morphology	Voltage range (V)	Cycles	Overpotential (V)	Ref. in the manuscript
Graphene nanosheets [unknown]	0.25 NaPF <sub>6</sub> DME <sup>1</sup>	9268 mAh g <sup>-1</sup> at 200 mA g <sup>-1</sup>	Na <sub>2</sub> O <sub>2</sub>	Film	1.5-4	10 (300 mA g <sup>-1</sup> to 1200 mAh g <sup>-1</sup> )	1.46 - 1.59	39
Nitrogen doped graphene [0.3;0.71 ]	0.5 mol dm <sup>-3</sup> NaSO <sub>3</sub> CF <sub>3</sub> DEGDME <sup>2</sup>	6000 mAh g <sup>-1</sup> or 3.63 mAh cm <sup>-2</sup> at 75 mA g <sup>-1</sup>	Na <sub>2</sub> O <sub>2</sub>	small particles	1.8-3.6	3 (75 mA g <sup>-1</sup> to 1150 mAh g <sup>-1</sup> )	0.6	40
Graphene nanosheets with Pt nanoparticles [unknown]	1M NaClO <sub>4</sub> :PC <sup>3</sup>	7574 mAh g <sup>-1</sup> at 0.1mA cm <sup>-2</sup>	Na <sub>2</sub> CO <sub>3</sub>	Nanomeric particles	2-3.5	10 (0.1 mA cm <sup>-2</sup> to 100 mAh g <sup>-1</sup> )	1.3	41
Ag-reduced graphene oxide [0.85 mg cm <sup>-2</sup> ]	1M NaPF <sub>6</sub> TEGDME <sup>3</sup>	566 mAh g <sup>-1</sup> at 0.1mA cm <sup>-2</sup>	NaO <sub>2</sub> , Na <sub>2</sub> O <sub>2</sub> and Na <sub>2</sub> O	Nanomeric particles	1.5-4.5	30 (0.2 mA cm <sup>-2</sup> to 0.125 mAh cm <sup>-2</sup> )	0.9	42
Reduced graphene oxide [0.15;0.5]	0.25M NaClO <sub>4</sub> DME <sup>3</sup>	40000 mAh g <sup>-1</sup> or 12 mAh cm <sup>-2</sup> at 0.1 mA cm <sup>-2</sup>	NaO <sub>2</sub>	Cubic particles	1.8-2.8	17 (0.1 mA cm <sup>-2</sup> to 1 mAh cm <sup>-2</sup> )	0.4	36
Micrometer-sized RuO <sub>2</sub> catalyst-coated B-rGO [0.5 mg cm <sup>-2</sup> ]	1M NaCF <sub>3</sub> SO <sub>3</sub> TEGDME <sup>3</sup>	0.5 mAh cm <sup>-2</sup> at 0.05mA cm <sup>-2</sup>	NaO <sub>2</sub>	Cubic particles	1.8-4.5	100 (0.05 mA cm <sup>-2</sup> to 0.5 mAh cm <sup>-2</sup> )	1	43
This work [1.5- 2.5,0.95]	0.1M NaClO <sub>4</sub> DME <sup>3</sup>	3594 mAh g <sup>-1</sup> or 6.61 mAh cm <sup>-2</sup> at 100 mA g <sup>-1</sup>	NaO <sub>2</sub>	Cubic particles	1.8-3.2	40 (100 mA g <sup>-1</sup> / 0.15-0.25 mA cm <sup>-2</sup> to 0.5 mAh cm <sup>-2</sup> ) 100 (100 mA g <sup>-1</sup> /0.15 mA cm <sup>-2</sup> to 0.15 mAh cm <sup>-2</sup> )	0.26	–

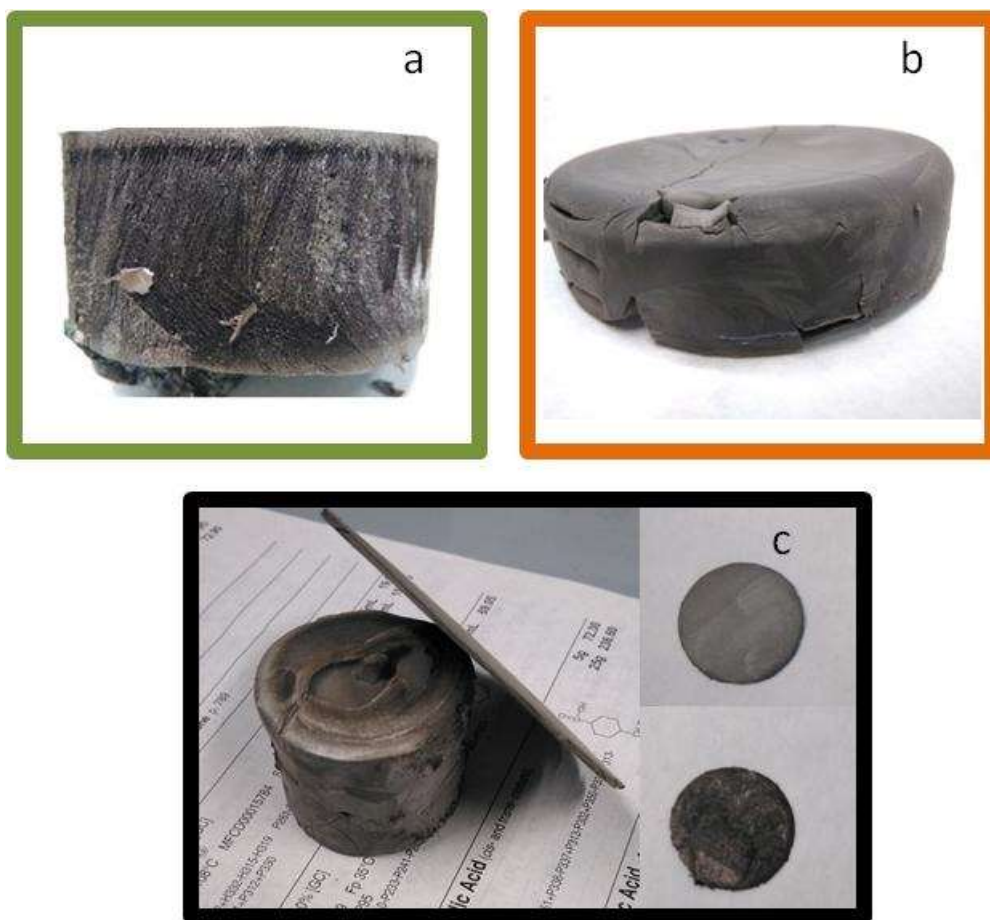
<sup>1</sup> dried air

<sup>2</sup> 1atm O<sub>2</sub>

<sup>3</sup> O<sub>2</sub>



**Fig S3. (a)** Voltage versus capacity of ArGO\_N graphene-based electrodes discharged galvanostatically at 100 mA g<sup>-1</sup> to full capacity in two electrolyte concentrations: 0.1 and 1 M NaClO<sub>4</sub> in DME. The difference in overpotential between 0.1 M and 1 M electrolyte solutions is indicated by an arrow.



**Fig. S4.** Images of the two prepared cryogels; (a) ArGO\_U and (b) ArGO\_N and (c) some self-standing binder-free electrodes processed from the aerogels.