Supporting Information.

Stacking-configuration-preserved Graphene Quantum Dots Electrochemically Obtained from CVD Graphene.

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S1. Survey XPS spectra and C/O content for GQDs.

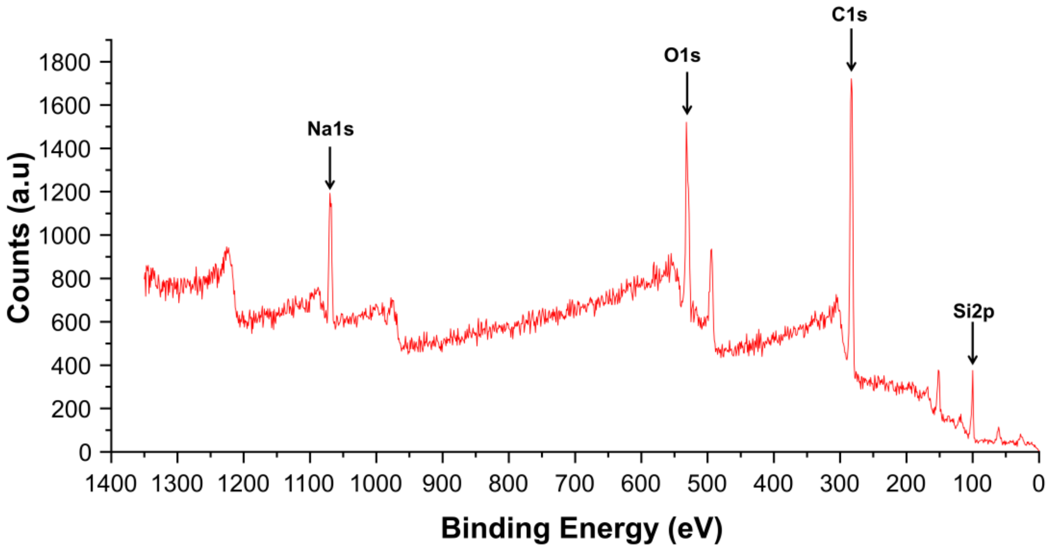


Table S1. XPS Element identification.

Name	Peak (eV)	FHWM (eV)	Area (CPS*eV)	Atomic %
C1s	285.55	4.66	129487.84	64.25
O1s	533.70	5.92	96033.76	19.72
Si2p	102.83	2.78	19495.26	9.63
Na1s	1071.93	4.88	62943.59	6.39

Figure S1. Survey XPS for GQDs. Table S1 shows element identification providing name, FHWM, peak area and atomic percentage.

S2. Corresponding electron diffraction simulation for NaO$_2$, Cu, CuO, Ni, and NiO.

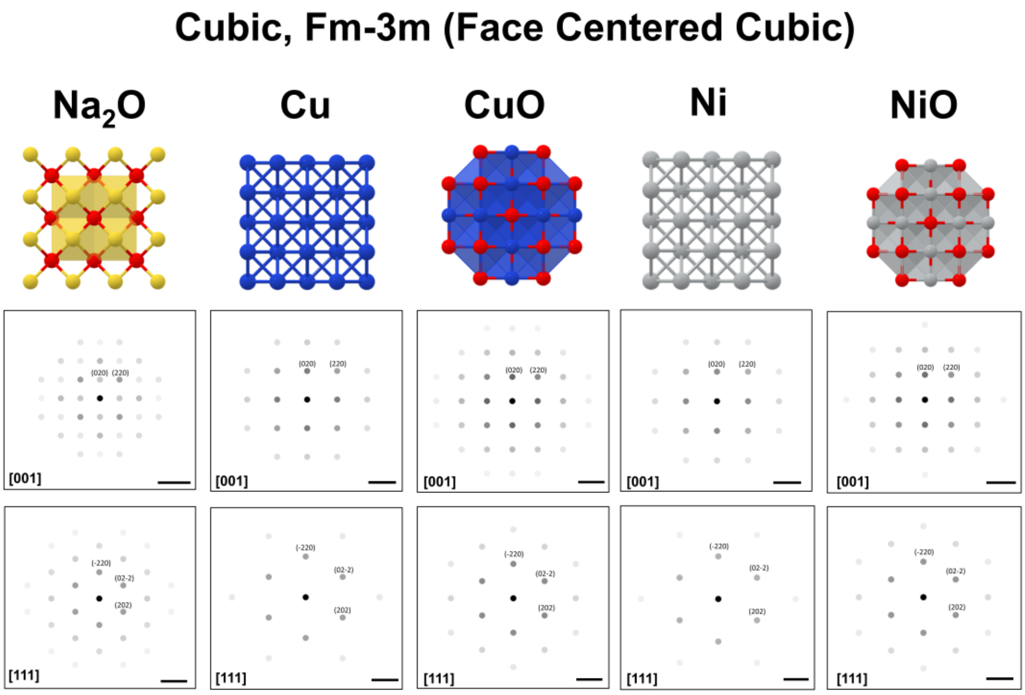


Table S2. Simulated electron diffraction hkl planes

Na$_2$O ($\dot{A}$)	Cu ($\dot{A}$)	CuO ($\dot{A}$)	Ni ($\dot{A}$)	NiO ($\dot{A}$)	(h,k,l)
3.167	2.065	2.444	2.06	2.417	111
2.742	1.789	2.117	1.738	2.093	200
1.939	1.265	1.497	1.229	1.48	220

Figure S2. Electron diffraction simulation for Na$_2$O, Cu, CuO, Ni, and NiO. Table S2 shows simulated electron diffraction hkl planes. Scale bar corresponds to 5 $\dot{A}^{-1}$.

S3. XRD for GQDs and NaOH.

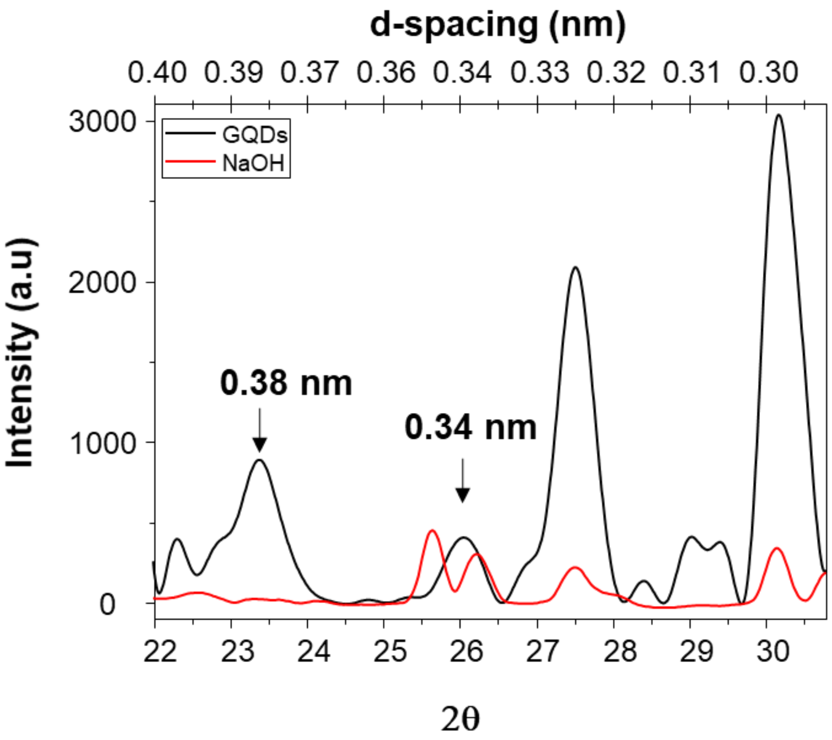


Figure S3. XRD spectra for as-prepared GQDs (black) and NaOH (red).

S4. Raman spectra for GQDs.

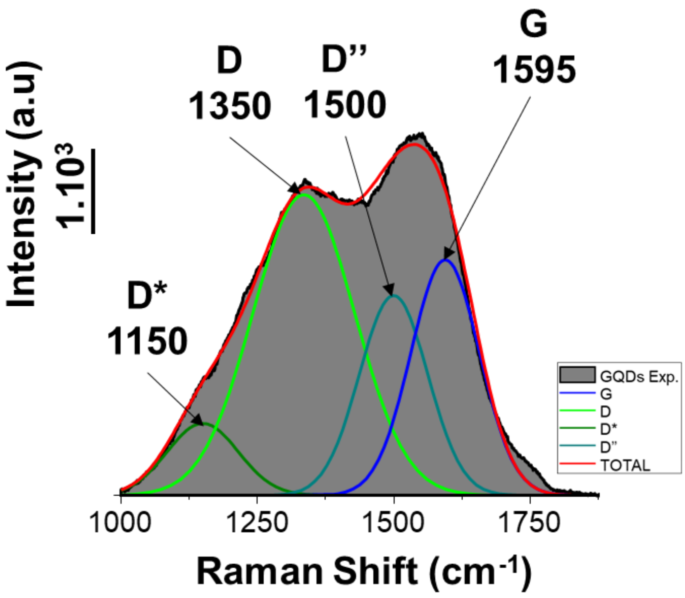


Figure S4. Raman spectra for as-synthesized GQDs.

S5. Additional AFM images and profiles of single and bilayer GQDs.

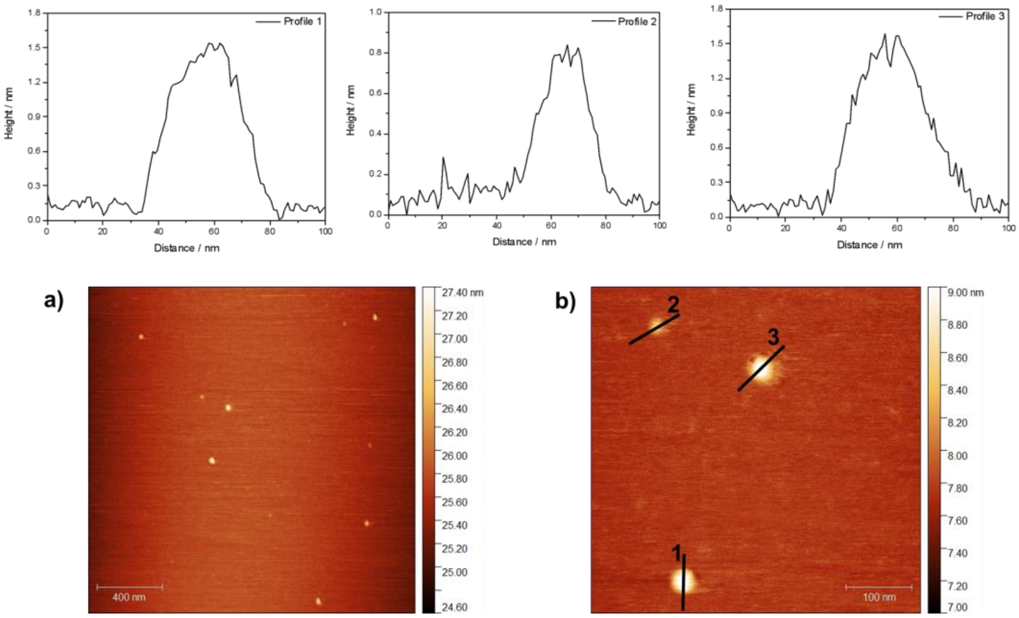
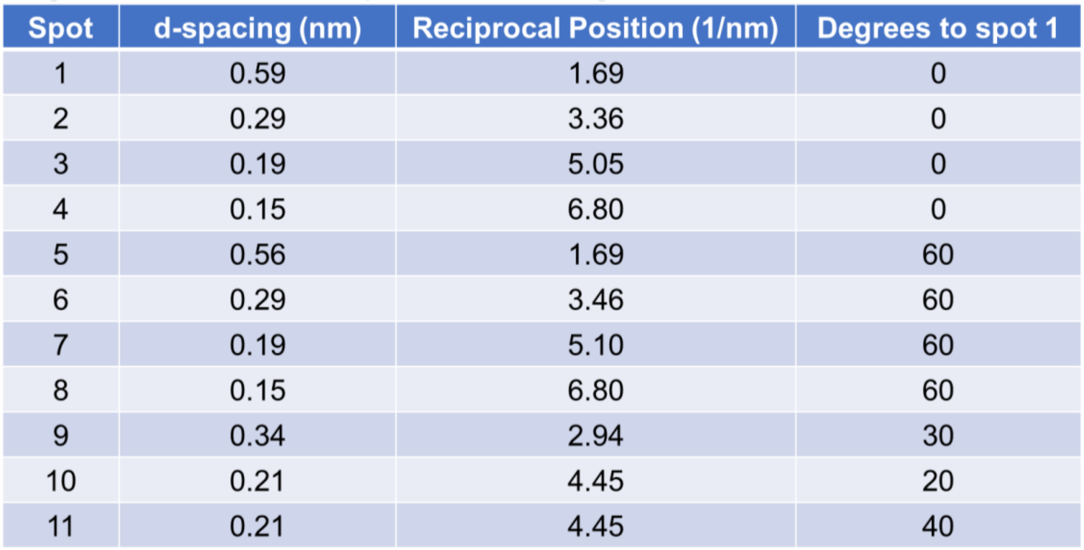


Figure S5. AFM images and profiles of bilayer and multi-layered GQDs.

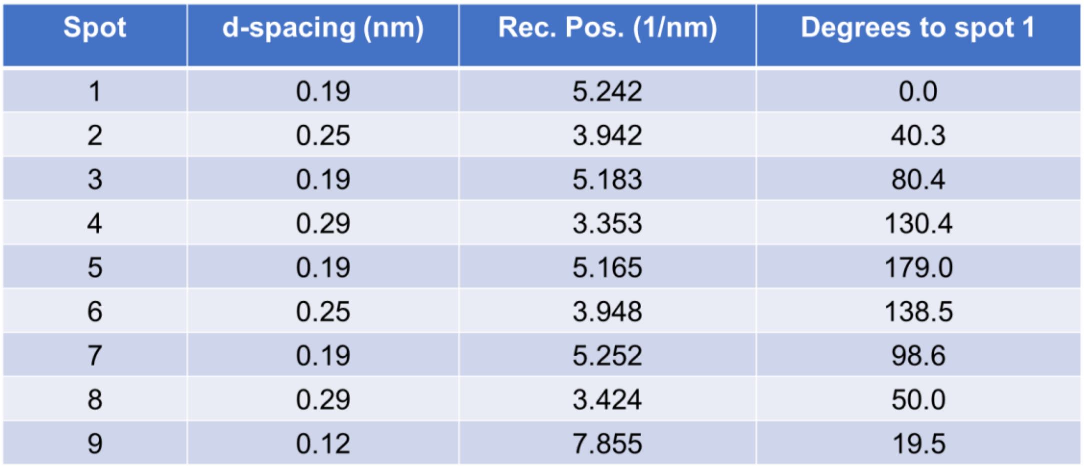
S6. The Table S3 shows particular information about the product evolved from the control experiment # 1 (CN1)



Spot	d-spacing (nm)	Reciprocal Position (1/nm)	Degrees to spot 1
1	0.59	1.69	0
2	0.29	3.36	0
3	0.19	5.05	0
4	0.15	6.80	0
5	0.56	1.69	60
6	0.29	3.46	60
7	0.19	5.10	60
8	0.15	6.80	60
9	0.34	2.94	30
10	0.21	4.45	20
11	0.21	4.45	40

Table S3. The Table shows the observed diffraction spots, their lattice spacing, and the angles between them for CN1 shown in Fig. 5 B in the main paper.

S7. The Table S4 shows particular information about the product evolved from the control experiment # 2 (CN2)



Spot	d-spacing (nm)	Rec. Pos. (1/nm)	Degrees to spot 1
1	0.19	5.242	0.0
2	0.25	3.942	40.3
3	0.19	5.183	80.4
4	0.29	3.353	130.4
5	0.19	5.165	179.0
6	0.25	3.948	138.5
7	0.19	5.252	98.6
8	0.29	3.424	50.0
9	0.12	7.855	19.5

Table S4. The Table shows the observed diffraction spots, their lattice spacing, and the angles between them for CN2 shown in Fig. 6 B in the main paper.

S8. Amorphous Carbon Nanoparticles and Multi-walled Carbon Nanotube obtained in Control #2 (CN2).

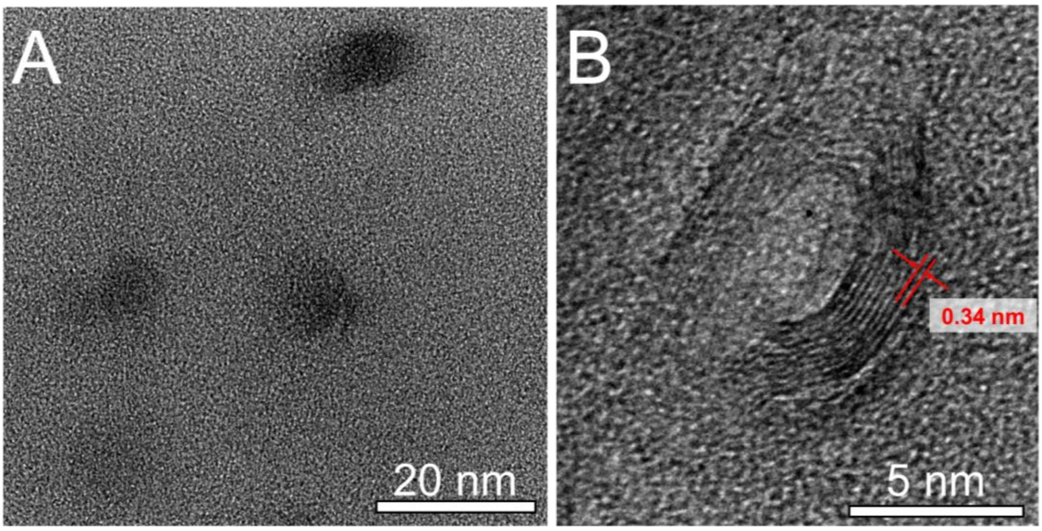


Figure S6. Amorphous Carbon Nanoparticles (A) and Multi-walled Carbon Nanotube (B) obtained in Control # 2 (CN2).

S9. Ethanol electro-oxidation in alkaline media with G/Ni Foams as WE.

Here we briefly study the electro-oxidation of ethanol in alkaline media using Graphene/Nickel Foam as working electrode and a platinum wire as counter with respect an Ag/AgCl reference electrode. Electro-oxidation of ethanol in alkaline media with nickel electrodes has been developed by Fleischmann and collaborators[1,2] around 1971-72, they laid down the foundation in our comprehension of this mechanism, extremely important for DEFC (Direct Ethanol Fuel Cells) and recently in the synthesis of graphene and carbon quantum dots in alkaline media. According to Fleischmann et al the electro-oxidation of simple chain alcohol in such circumstances it's a 4 e^- mediated process, this process involves the formation of oxides in the working electrode as a reversible process and some radical intermediates.

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1) $OH^- + lower\ oxide \underset{fast\rightarrow}{\Longleftrightarrow} higher\ oxide + H_2O + e^-$

2) $(organic\ substrate)_{sol} \underset{on\ higher\ oxide}{\Longleftrightarrow} (organic\ substrate)_{ads}$

3) $Higher\ Oxide + (Organic\ Substrate\ )_{ads} \overset{*}{\Rightarrow} lower\ oxide + radical\ intermediate$

$*\ rate\ determining\ hydrogen\ extraction$

4) $Radical\ Intermediate + (n-1)higher\ oxide$
$\underset{yields}{\Longrightarrow} (n-1)lower\ oxide + product$

This is a general route for the electro-oxidation of simple chain alcohols in alkaline media, with different metal catalyst. In recent years Barbosa et al[3] take this same approach and using an FTIR they determined the different participants involved in the electro-oxidation of ethanol on polycrystalline nickel in alkaline media; according to their findings we can establish:

Lower oxide = Ni $(OH)_2$ = Nickel Oxide (II)

Higher Oxide = NiO (OH) = Nickel oxide hydroxide

Organic Substrate = CH_3CH_2OH = Ethanol

Radical Intermediate = CH_3CHO = Acetaldehyde

Product = CH_3COOH = Acetic Acid (Acetate for pH>10)

Yielding the next 5 equations for the electrode dynamics:

1) $OH^- + \mathrm{Ni(OH)2} \underset{fast\rightarrow}{\Longleftrightarrow} \mathrm{NiO(OH)} + H_2O + e^-$

2) $(\mathrm{CH_3CH_2OH})_{sol} \underset{on\ higher\ oxide}{\Longleftrightarrow} (\mathrm{CH_3CH_2OH})_{ads}$

3) $\mathrm{NiO(OH)} + (\mathrm{CH_3CH_2OH}\ )_{ads} \overset{*}{\Rightarrow} \mathrm{Ni(OH)2} + \mathrm{CH_3CHO}$

$* \, rate \; determining \; hydrogen \; extraction$

4) $$\mathrm{CH_3CHO} + (n-1)\mathrm{NiO(OH)} \underset{yields}{\Longrightarrow} (n-1)\mathrm{Ni(OH)2} + \mathrm{CH_3COOH}$$

5) $$\mathrm{CH_3COOH} + \mathrm{OH^-} \underset{yields}{\Longrightarrow} \mathrm{CH_3COO^-}$$

Now we can establish a direct correlation between experimental data and the theory behind ethanol electro-oxidation, for that purpose we perform different cycle voltagramm in a 3 electrode set -up for the electro-oxidation of ethanol. Figure S1A shows a schematic of the experimental set-up. Graphene/Nickel (Nickel) Foam is used as a working electrode and a platinum wire as counter electrode. The reference voltage is set trough an Ag/AgCl reference electrode. Figure S7B shows a cyclovoltammetry measurements of ethanol electrooxidation on Graphene/Nickel (Nickel) foams. Control group without ethanol is added for completeness. Ethanol can be electro oxidized to Acetate in a 0.55-0.85 V window with respect to an Ag/AgCl reference electrode on a G/Ni or Ni foam as a working electrode. The presence of Nickel Oxides is critical to the electro-oxidation of ethanol because these oxides are participants in all the redox reactions. The incorporation of Graphene is enhancing the amount of ethanol oxidized but shifts the over potential for electro-oxidation.

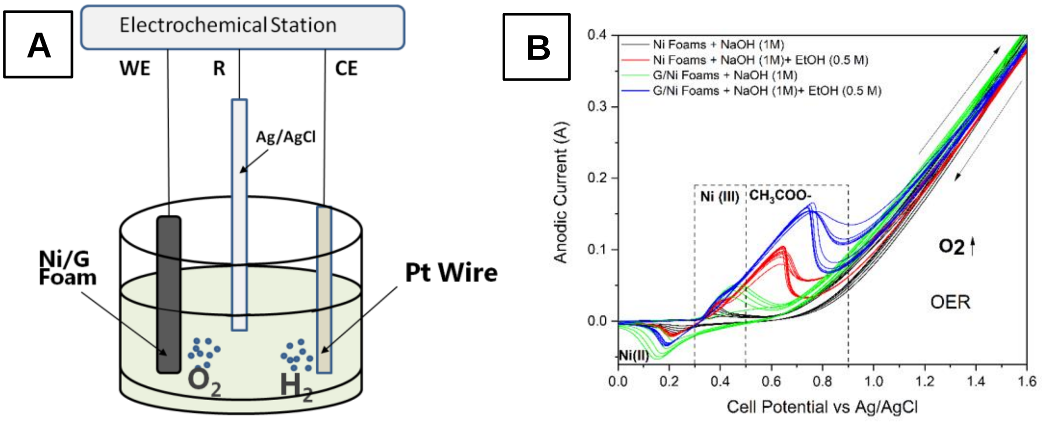

Figure S7. (a) Scheme showing 3-electrode set-up electrochemical station comprised of graphene grown on Ni foam (anode), Pt as a cathode, and Ag/AgCl reference electrode immersed in 1.0 M NaOH and 0.5 M ethanol (A). Cyclic voltagramm of graphene grown on Nickel (Nickel) Foam and the corresponding control experiments in the absence of graphene and ethanol.

References.

(1) Fleischmann, M.; Korinek, K.; Pletcher, D. The Oxidation of Organic Compounds at a Nickel Anode in Alkaline Solution. *J. Electroanal. Chem. Interfacial Electrochem.* **1971**, *31* (1), 39–49. https://doi.org/10.1016/S0022-0728(71)80040-2.

(2) Fleischmann, M.; Korinek, K.; Pletcher, D. The Kinetics and Mechanism of the Oxidation of Amines and Alcohols at Oxide-Covered Nickel, Silver, Copper, and Cobalt Electrodes. *J. Chem. Soc. Perkin Trans. 2* **1972**, No. 10, 1396–1403. https://doi.org/10.1039/P29720001396.

(3) Barbosa, A. F. B.; Oliveira, V. L.; van Drunen, J.; Tremiliosi-Filho, G. Ethanol Electro-Oxidation Reaction Using a Polycrystalline Nickel Electrode in Alkaline Media: Temperature Influence and Reaction Mechanism. *J. Electroanal. Chem.* **2015**, *746*, 31–38. https://doi.org/10.1016/j.jelechem.2015.03.024.