

## Electronic Supplementary Information

# Simple Coordination Complex-Derived Three-Dimensional Mesoporous Graphene as an Efficient Bifunctional Oxygen Electrocatalyst

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## 1. Materials and Methods

All chemicals and solvents were of reagent grade and were used without further purification. Infrared spectra were recorded using a ThermoFisher Scientific iS10 FT-IR spectrometer. Raman spectroscopy measurements were performed using a micro-Raman system (WITec) with an excitation energy of 2.41 eV (532 nm). Elemental analyses were performed at the UNIST Central Research Facilities Center (UCRF) at Ulsan National Institute of Science and Technology (UNIST). TGA was performed under N<sub>2</sub>(g) at a scan rate of 10 °C min<sup>-1</sup>, using a TGA Q50 from TA instruments. X-ray powder diffraction data were recorded on a Bruker D2 phaser diffractometer at 30 kV and 10 mA for Cu K $\alpha$  ( $\lambda$  = 1.54050 Å), with a step size of 0.02° in 2 $\theta$ . N<sub>2</sub> sorption isotherms of *meso*G/Ni and 3D *meso*G were obtained using a BELSORP-max at 77 K. Prior to the adsorption measurements, the samples were evacuated ( $p < 10^{-5}$  mbar) at 100 °C for 4 h. The specific surface area was determined in the relative pressure range from 0.05 to 0.3 of the Brunauer-Emmett-Teller (BET) plot, and the total pore volume was calculated from the amount adsorbed at a relative pressure of about 0.98-0.99. TEM images were obtained using a JEOL JEM-2100F microscope. AR-TEM images were collected using an image-side spherical aberration corrected TEM (Titan<sup>3</sup> G2 60-300, FEI Company, Netherlands), operated at 80 kV.

### 1.1. Synthesis

**Preparation of [Ni<sub>2</sub>(EDTA)].** A DMF (20 mL) solution of Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (1.16 g, 4.0 mmol) was added to a DMF (30 mL) solution of H<sub>4</sub>EDTA (0.59 g, 2.0 mmol) and triethylamine (1.5 mL, 10.8 mmol). A precipitate formed soon after the two solutions were mixed. The resulting solid was filtered and washed with neat DMF. After drying in vacuo at room temperature overnight, a blue powder was obtained. Yield: 1.15 g (95%). IR (KBr):  $\nu =$

3373 (br;  $\nu(\text{O-H})$ ), 2975 (w;  $\nu(\text{aliphatic C-H})$ ), 2940 (w;  $\nu(\text{aliphatic C-H})$ ), 1660 (s;  $\nu(\text{C=O, DMF})$ ), 1590 (s;  $\nu_{\text{as}}(\text{carboxylate})$ ), 1403 (s;  $\nu_{\text{s}}(\text{carboxylate})$ ), 482  $\text{cm}^{-1}$  (w;  $\nu(\text{Ni-N})$ ). Anal. Calcd for  $[\text{Ni}_2(\text{EDTA})]\cdot 1.5\text{DMF}\cdot 5\text{H}_2\text{O}$ ,  $\text{Ni}_2\text{C}_{14.5}\text{H}_{32.5}\text{N}_{3.5}\text{O}_{14.5}$ : C 28.77, H 5.41, N 8.10; found: C 28.92, H 4.26, N 8.06.

**Thermolysis.**  $[\text{Ni}_2(\text{EDTA})]$  (1.15 g) was ground into a fine powder and heated at  $10\text{ }^\circ\text{C min}^{-1}$  under a nitrogen flow rate of  $500\text{ mL min}^{-1}$ . After the temperature reached  $1000\text{ }^\circ\text{C}$ , the material was maintained at that temperature for 1 h. After cooling to room temperature rapidly, 0.32 g of a black powder (*mesoG/Ni*) was obtained. Anal. Found for *mesoG/Ni*: C 33.37, H 0.13, N 0.72, O 1.23 (Table S1).

**Etching method.** *mesoG/Ni* composites obtained after the thermolysis of  $[\text{Ni}_2(\text{EDTA})]$  were etched according to the following procedure to obtain pure 3D *mesoG* materials<sup>1</sup>: 1 M  $\text{FeCl}_3\cdot 6\text{H}_2\text{O}$  (8 mL) and 1 M HCl (8 mL) were added to a vial containing the *mesoG/Ni* (0.32 g). The mixture was heated at  $80\text{ }^\circ\text{C}$  for 3 h. After cooling to room temperature, the solid product was collected by centrifugation at 7000 rpm and was washed with distilled water several times. The product was dried in vacuo at room temperature overnight to yield a black powder (3D *mesoG*, 0.10 g). Anal. Found for 3D *mesoG*: C 91.63, H 0.35, N 1.71, O 2.22 (Table S1).

## 1.2 Electrochemical Measurements

Electrochemical characterization of the catalyst was carried out using a high-power potentiostat (IviumStat, Ivium Technologies, The Netherlands). Electrode rotation and gas purging were performed by a rotating ring disk electrode rotator (RRDE-3A, ALS, Japan). The electrochemical experiments were performed at room temperature ( $23\sim 25\text{ }^\circ\text{C}$ ) using a three-compartment electrochemical cell. Deionized water and ultra-high purity KOH (99.99%,

Aldrich) were used for the electrochemical characterization. A Pt-wire (for noble metal catalysts) or graphite rod (for 3D *mesoG*) was used as the counter electrode, and Hg/HgO (CHI152, CH instruments, USA) filled with 1 M KOH was used as a reference electrode. In this report, all potentials were corrected with respect to the reversible hydrogen electrode (RHE). The Hg/HgO reference electrode was calibrated with respect to the RHE on a daily basis. As such, a two-compartment electrochemical cell was built where the RHE (HydroFlex, Gaskatel GmbH, Germany) and Hg/HgO were used as the working electrode and reference electrode, respectively. The electrodes were immersed in N<sub>2</sub>-saturated 0.1 M KOH until the open circuit voltage (OCV) was stabilized. After at least 30 min, the resulting OCV was used as the calibration value. The calibration result was  $E(\text{RHE}) = E(\text{Hg/HgO}) + 0.884 (\pm 0.001)$  V.

Pt/C (20 wt% Pt, HiSPEC-3000, Johnson-Matthey) and Ir/C (20 wt% Ir, Premetek) were used as benchmarks for catalytic activity. The ink for the noble metal catalysts was composed of 5 mg of catalyst, 0.1 mL of DI water, 0.04 mL of 5 wt% Nafion (Aldrich), and 1.06 mL of anhydrous ethanol (Samchun chemical). The 3D *mesoG* catalyst ink consisted of 30 mg of catalyst, 0.1 mL of DI water, 0.08 mL of the Nafion, and 0.98 mL of anhydrous ethanol, resulting in the catalysts concentration to be 25 mg/mL. The catalyst slurry was ultrasonicated for at least 30 min. The rotating disk electrode (RDE, ALS) was polished on a polishing pad with a 0.3  $\mu\text{m}$  alumina suspension prior to ink deposition, generating a mirror finish. After trace alumina was removed, 3  $\mu\text{L}$  of the homogeneous ink was dropped onto the glassy carbon electrode (4 mm in diameter) of the RDE using a micro-syringe (Hamilton). The ink was dried at 70  $^{\circ}\text{C}$  for 2 min under static conditions. The catalyst loading was 0.6  $\text{mg cm}^{-2}$  for the 3D *mesoG* and 0.1  $\text{mg cm}^{-2}$  for Pt/C and Ir/C (0.02  $\text{mg}_{\text{metal}} \text{cm}^{-2}$ ).

For *mesoG*, the air-bubbles generated on the catalyst layer should be removed before the electrochemical measurements by applying a constant potential at 0.05 V (vs. RHE) with an RDE rotation speed of 1600 rpm with concomitant O<sub>2</sub> bubbling. Electrochemical cleaning of the catalyst surface was conducted for the noble metal catalysts. Fifty potential cycles from 0.05 to 1.20 V (vs. RHE) were applied in N<sub>2</sub>-saturated 0.1 M KOH at a potential scan rate of 500 mV s<sup>-1</sup>. To measure the OER activity, cyclic voltammetry (CV) was carried out from 1.20 to 1.70 V (vs. RHE) at a scan rate of 20 mV s<sup>-1</sup> (for Pt/C catalyst, a potential cycle from 1.20 to 2.00 V (vs. RHE) was applied to obtain an overpotential at 10 mA cm<sup>-2</sup>). The cathodic and anodic currents in the CV curves were displayed after being averaged and iR-corrected using series resistance (R<sub>s</sub>). To obtain the R<sub>s</sub> value, electrochemical impedance spectroscopy was carried out at the open circuit potential in O<sub>2</sub>-saturated electrolyte. The R<sub>s</sub> was determined at a high frequency range, and the average R<sub>s</sub> was 55 Ω. To compare the stability of the catalysts in the OER, the CV was scanned from 1.20 to 1.70 V 100 times at 20 mV s<sup>-1</sup>, and the initial and final current densities at 1.7 V were compared (Figure 3).

The ORR activity of the noble metal catalyst was measured in a potential profile (-0.01 → 1.1 V) at a scan rate of 20 mV s<sup>-1</sup> with an electrode rotation of 1600 rpm in an O<sub>2</sub>-saturated electrolyte. Then, the background current measured in N<sub>2</sub>-saturated electrolyte was subtracted. The LSV of the third measurement was displayed in this report. The ORR activity of 3D *mesoG* was measured in a potential profile (1.1 → 0.2 V) at 5 mV s<sup>-1</sup>. Because the resulting linear sweep voltammogram (LSV) of the 3D *mesoG* can include capacitive current due to high carbon loading, additional LSVs were obtained to correct for the background current in the N<sub>2</sub>-saturated solution before or after the ORR measurements. The current density after the background current correction was reported.

Tafel plots were obtained from the following Tafel equation to compare the ORR and the OER kinetics.

$$E_{\text{applied}} = b \log(j_K) + C$$

Here,  $E_{\text{applied}}$ ,  $b$ ,  $j_K$ , and  $C$  are applied potential ( $iR$ -corrected), Tafel slope, kinetic current, and constant, respectively. For the OER Tafel plot, the obtained current density was used as the kinetic current. For the ORR Tafel plot, the mass-transfer current should be corrected according to the Koutecky-Levich equation.

$$\frac{1}{j_K} = \frac{j \times j_L}{j_L - j}$$

Here,  $j$  and  $j_L$  are the measured current and diffusion-limited current densities, respectively.

**Table S1.** Elemental composition of [Ni<sub>2</sub>(EDTA)], *meso*G/Ni, and 3D *meso*G.

| Elements | Materials  | [Ni <sub>2</sub> (EDTA)] <sup>a</sup> | <i>meso</i> G/Ni       | 3D <i>meso</i> G       |
|----------|------------|---------------------------------------|------------------------|------------------------|
|          |            | (1.15 g) <sup>b</sup>                 | (0.32 g) <sup>b</sup>  | (0.10 g) <sup>b</sup>  |
| Ni       | wt%        | 19.4                                  | 60.8 <sup>c</sup>      | 1.38 <sup>c</sup>      |
|          | Weight (g) | 0.22                                  | 0.20                   | 1.4 × 10 <sup>-3</sup> |
| C        | wt%        | 28.77                                 | 33.37                  | 91.63                  |
|          | Weight (g) | 0.33                                  | 0.11                   | 0.091                  |
| H        | wt%        | 5.41                                  | 0.13                   | 0.35                   |
|          | Weight (g) | 0.062                                 | 4.2 × 10 <sup>-4</sup> | 3.5 × 10 <sup>-4</sup> |
| N        | wt%        | 8.10                                  | 0.72                   | 1.71                   |
|          | Weight (g) | 0.093                                 | 2.3 × 10 <sup>-3</sup> | 1.7 × 10 <sup>-3</sup> |
| O        | wt%        | 38.33                                 | 1.23                   | 2.22                   |
|          | Weight (g) | 0.44                                  | 3.9 × 10 <sup>-3</sup> | 2.2 × 10 <sup>-3</sup> |

<sup>a</sup> Calculated values based on the formula determined by elemental combustion analysis.

<sup>b</sup> Weight of the sample obtained by experiments mentioned in methods section.

<sup>c</sup> Calculated values from the amount of NiO, confirmed from TGA results obtained under an oxygen atmosphere (see Figure S2 and S3).

**Table S2.** Comparison of Raman data of graphene- and CNT-related materials.

| Materials                                            | Peak position (cm <sup>-1</sup> ) |       |       | Ratio<br>$I_D/I_G$ | References          |
|------------------------------------------------------|-----------------------------------|-------|-------|--------------------|---------------------|
|                                                      | D                                 | G     | 2D    |                    |                     |
| Graphene oxide (GO)                                  |                                   |       |       | 1.88               | 2                   |
| Graphene aerogel (GA)                                | ~1325                             | ~1590 | ~2645 | 2.31               |                     |
| Graphene oxide aerogel (GOA)                         |                                   |       |       | 2.49               |                     |
| Reduced graphene oxide (rGO)                         | 1337                              | 1588  | -     | 1.30               | 3                   |
| Hydrazine-reduced GO                                 | 1349                              | 1577  | -     | 0.74               | 4                   |
| e <sup>-</sup> beam-reduced GO                       | 1328                              | 1594  | -     | 1.51               |                     |
| GO                                                   | 1358                              | 1594  | 2680  | 0.77               | 5                   |
| rGO <sub>HI-AcOH</sub>                               | 1350                              | 1581  | -     | 1.10               |                     |
| rGO/CNT                                              | 1353                              | 1582  | 2697  | 1.04               | 6                   |
| Repaired GO with multi-layered graphene balls (RGGB) | 1355                              | 1575  | 2700  | 0.91               | 7                   |
| Multilayer graphene balls                            | 1340                              | 1580  | 2650  | > 1                | 8                   |
| Film of MWCNTs                                       | 1349                              | 1579  | 2701  | 0.85               | 9                   |
| MWCNTs                                               | 1354                              | 1584  | 2709  | 0.70               | 10                  |
| Aligned carbon nanotubes                             | 1348                              | 1581  | 2697  | 1.3                | 11                  |
| <b>3D mesoG from [Ni<sub>2</sub>(EDTA)]</b>          | 1340                              | 1575  | 2676  | 0.70               | <i>Present work</i> |

**Table S3.** Potentials required to reach 10 mA cm<sup>-2</sup> for OER and -3 mA cm<sup>-2</sup> for ORR, and the potential difference to derive 10 mA cm<sup>-2</sup> and -3 mA cm<sup>-2</sup>.

| Catalysts        | $E_{OER}$                                 | $E_{ORR}$                                 | $E_{OER} - E_{ORR}$ |
|------------------|-------------------------------------------|-------------------------------------------|---------------------|
|                  | @ 10 mA cm <sup>-2</sup> (V) <sup>a</sup> | @ -3 mA cm <sup>-2</sup> (V) <sup>b</sup> | (V)                 |
| 3D <i>meso</i> G | 1.56                                      | 0.76                                      | 0.80                |
| Ir/C             | 1.58                                      | 0.70                                      | 0.88                |
| Pt/C             | 1.83                                      | 0.88                                      | 0.95                |

<sup>a</sup> 10 mA cm<sup>-2</sup> is the current density required for photochemical fuel production with a 10% solar-to-fuel efficiency at AM1.5G.

<sup>b</sup> -3 mA cm<sup>-2</sup> corresponds to roughly half of the diffusion-limited current density during the ORR at a rotation speed of 1600 rpm.

**Table S4.** Comparison of ORR and OER activity parameters with reported carbon-based bifunctional electrocatalysts. Potentials to reach 10

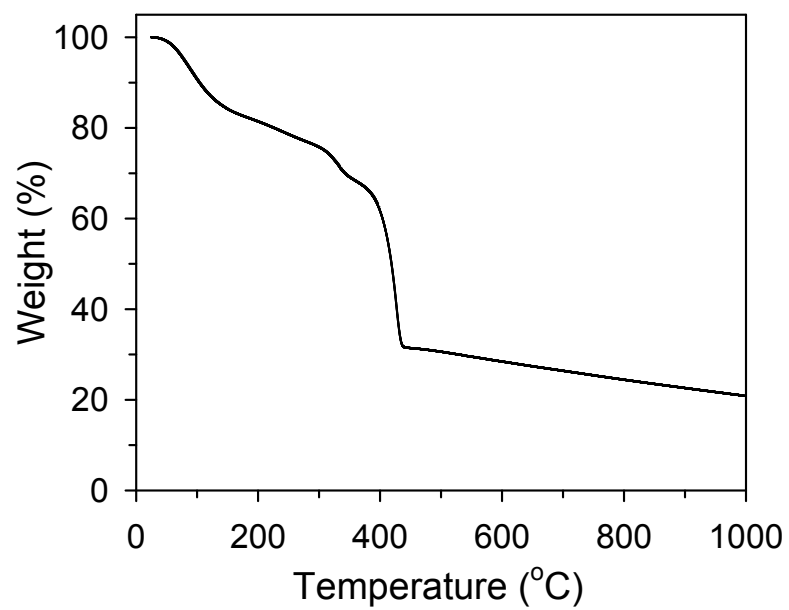
| Catalyst                                  | Catalyst Loading (mg cm <sup>-2</sup> ) | Electrolyte      | OER                                 |                                       | ORR                                 |             | Oxygen Electrode ( $E_{OER} - E_{ORR}$ ) | References                 |
|-------------------------------------------|-----------------------------------------|------------------|-------------------------------------|---------------------------------------|-------------------------------------|-------------|------------------------------------------|----------------------------|
|                                           |                                         |                  | Tafel Slope (mV dec <sup>-1</sup> ) | $E_{OER}$ (@ 10 mA cm <sup>-2</sup> ) | Tafel Slope (mV dec <sup>-1</sup> ) | $E_{ORR}^a$ |                                          |                            |
| <b>3D mesoG</b>                           | <b>0.6</b>                              | <b>0.1 M KOH</b> | <b>93<sup>b</sup></b>               | <b>1.56</b>                           | <b>34<sup>b</sup></b>               | <b>0.76</b> | <b>0.80</b>                              | <b><i>Present Work</i></b> |
| N-doped graphitic carbon                  | 0.2                                     | 0.1 M KOH        | ~110                                | 1.61                                  | -                                   | 0.77        | 0.84                                     | 12                         |
| N-doped graphene-1000                     | 0.14                                    | 0.1 M KOH        | -                                   | 1.76 (@ 2 mA cm <sup>-2</sup> )       | -                                   | 0.72        | >1.04                                    | 13                         |
| Thermally reduced GO/NCNT                 | 0.41                                    | 0.1 M KOH        | -                                   | 1.75                                  | -                                   | 0.87        | 0.88                                     | 14                         |
| N-doped graphene/CNT                      | 0.25                                    | 0.1 M KOH        | 83                                  | 1.63                                  | -                                   | 0.63        | 1.00                                     | 15                         |
| N-doped graphene/CNT                      | 0.2                                     | 0.1 M KOH        | -                                   | 1.65                                  | -                                   | 0.74        | 0.91                                     | 16                         |
| CNT@NCNT                                  | 0.25                                    | 0.1 M KOH        | -                                   | 1.76                                  | -                                   | 0.70        | 1.06                                     | 17                         |
| S/N_Fe-27                                 | 0.8                                     | 0.1 M KOH        | -                                   | 1.78                                  | -                                   | 0.87        | 0.91                                     | 18                         |
| P-doped carbon nitride-carbon fiber paper | ~0.20                                   | 0.1 M KOH        | 61.6                                | 1.63                                  | 122                                 | 0.67        | 0.96                                     | 19                         |

mA cm<sup>-2</sup> for OER and -3 mA cm<sup>-2</sup> for ORR, and oxygen electrode values are summarized.

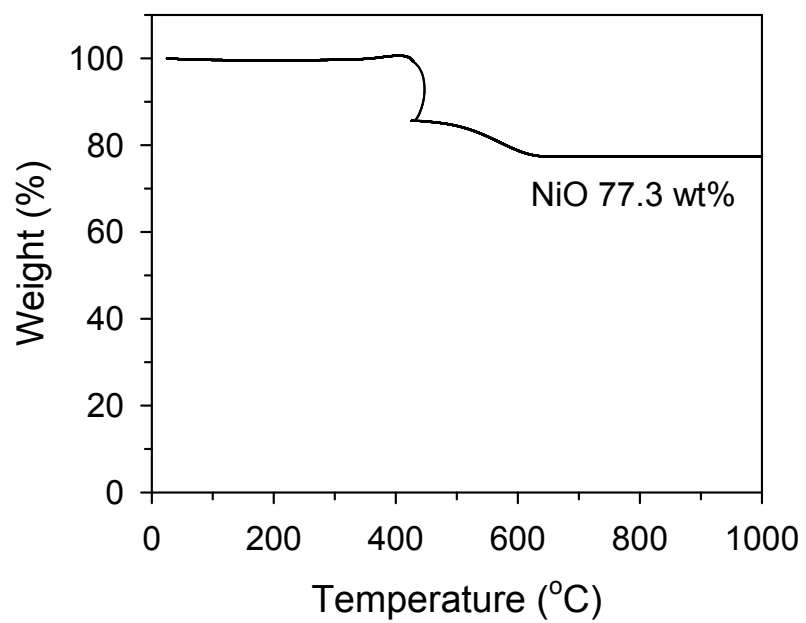
<sup>a</sup>  $E_{ORR}$  is determined as half-wave potential due to the different rotation speeds.

<sup>b</sup> Tafel plots for 3D mesoG are presented in Figure S8.

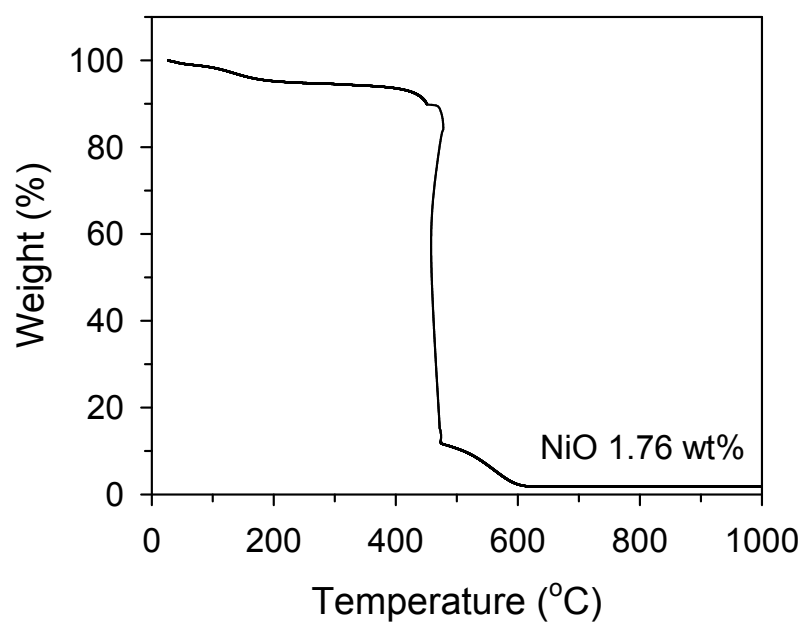




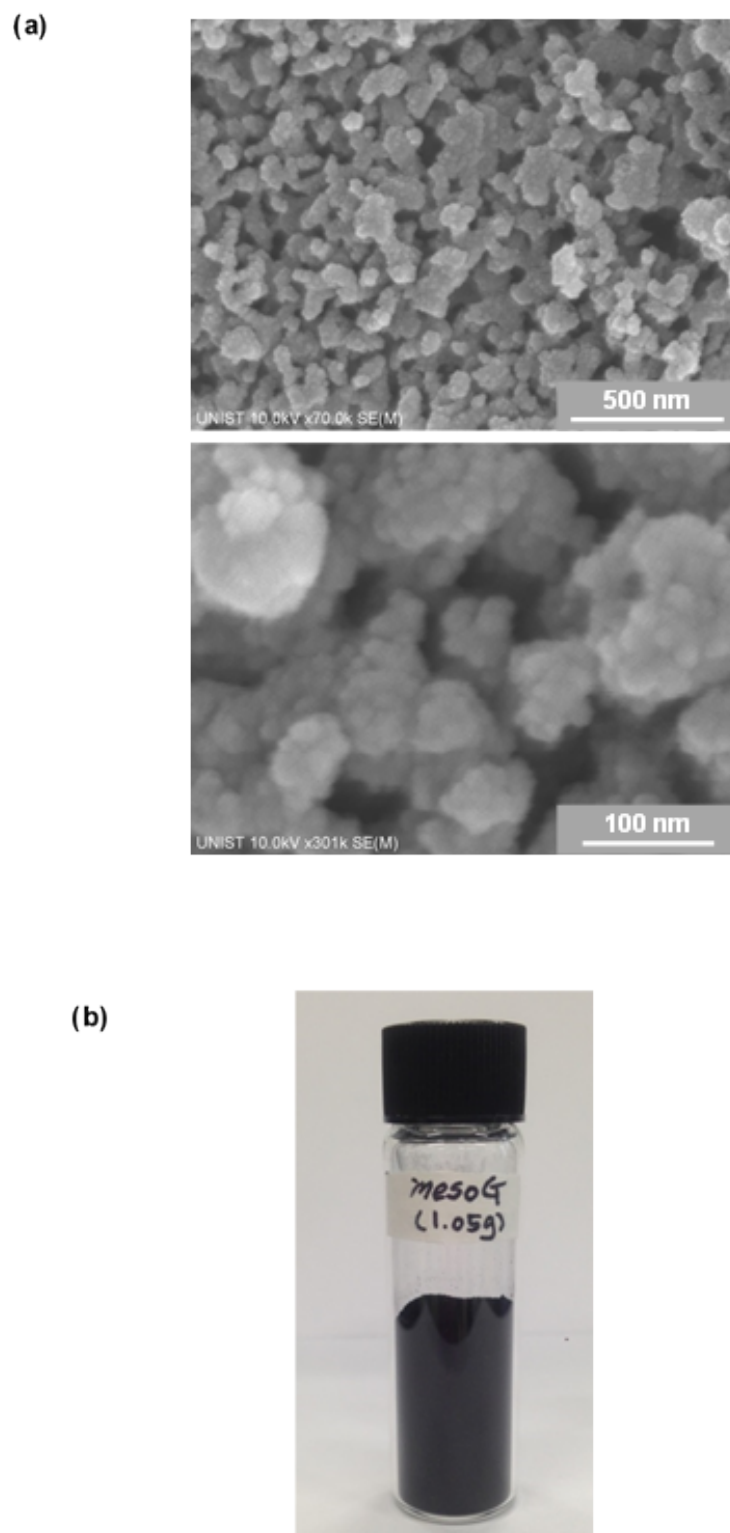
**Figure S1.** TGA trace of  $[\text{Ni}_2(\text{EDTA})]$  obtained under a nitrogen atmosphere.



**Figure S2.** TGA trace of *mesoG*/Ni obtained under an oxygen atmosphere. From the amount of the resultant solid (NiO), the Ni content in *mesoG*/Ni was calculated to be 60.8 wt%.

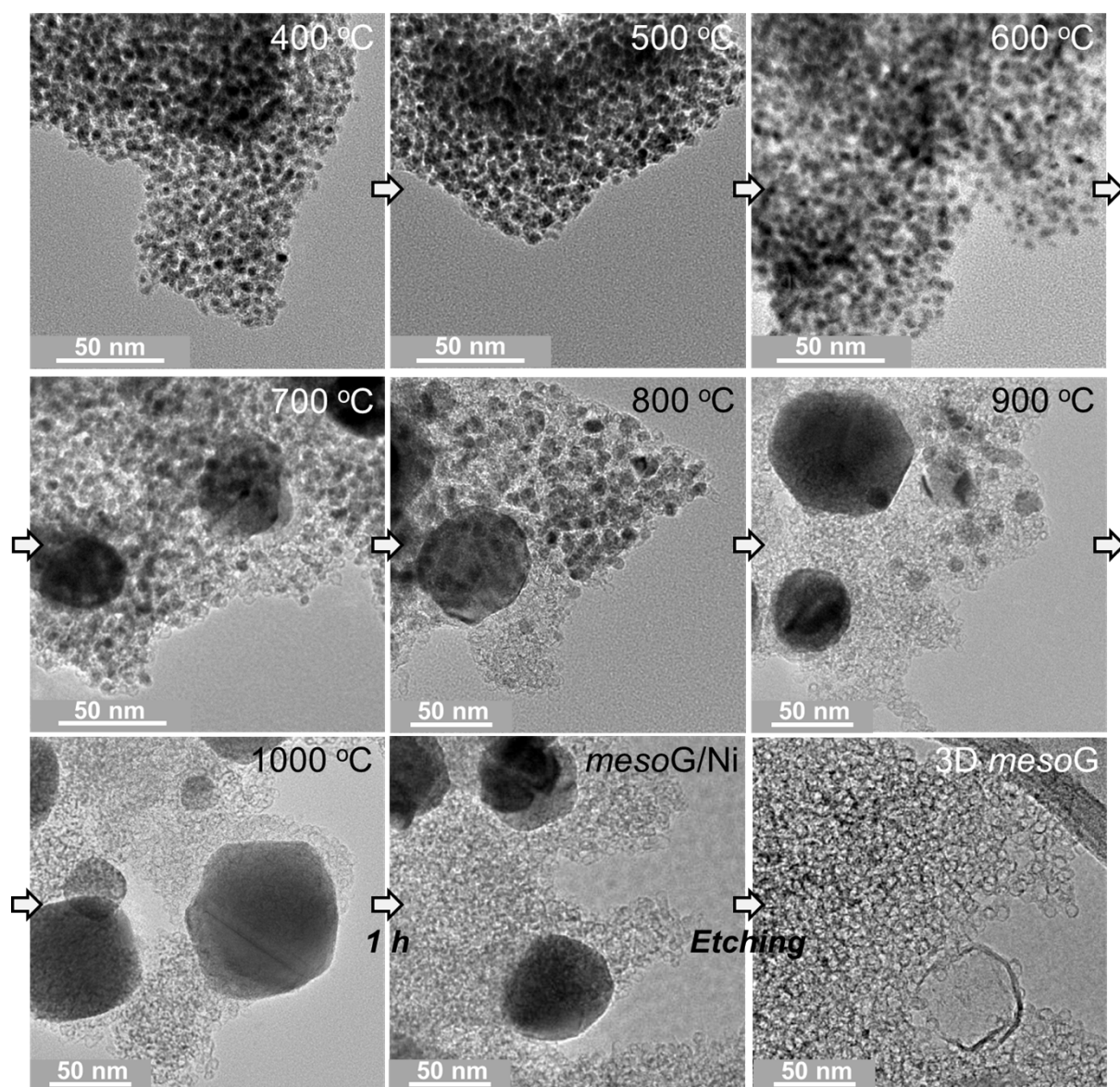


**Figure S3.** TGA trace of 3D *mesoG* obtained under an oxygen atmosphere. From the amount of the resultant solid (NiO), the Ni content in 3D *mesoG* was calculated to be 1.38 wt%.

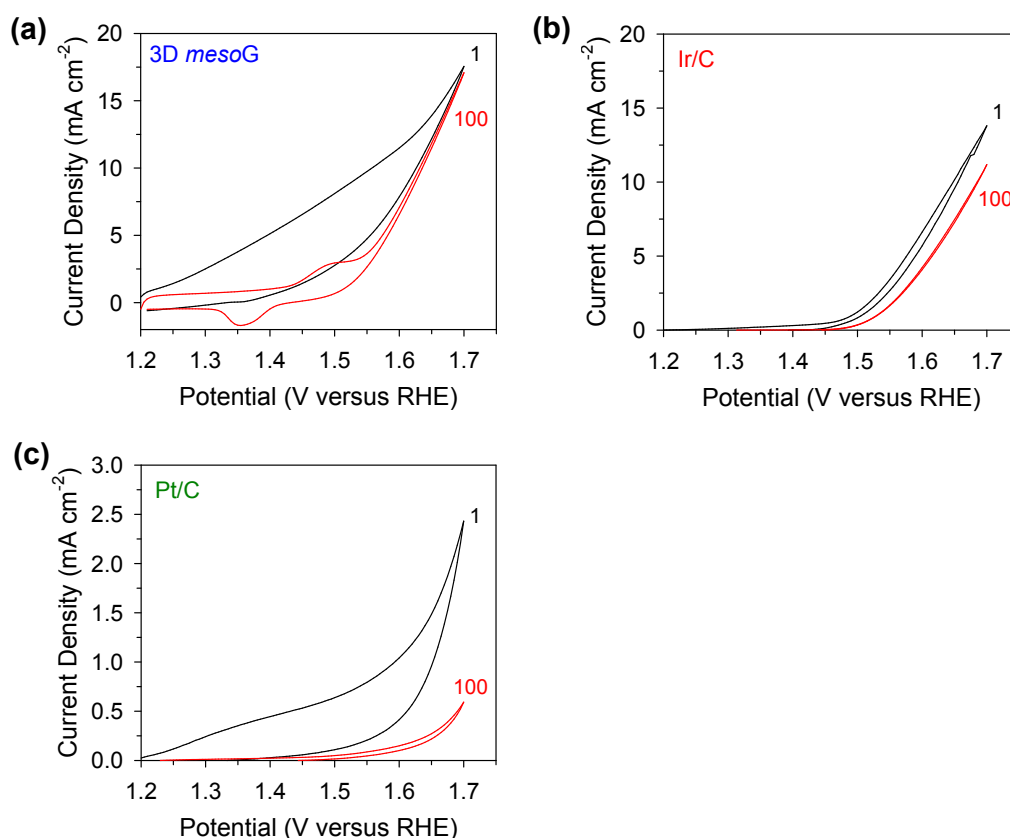


**Figure S4.** Micro- and macro-scope analyses. (a) SEM images of the 3D *mesoG*. (b) Photographic images of the 3D *mesoG* (1.05 g).

**Figure S5.** Temperature-dependent evolution of mesoporous graphene structures during thermal conversion of  $[\text{Ni}_2(\text{EDTA})]$  to *mesoG*/Ni. (a) VT-XRPD data for  $[\text{Ni}_2(\text{EDTA})]$ . (b) Size of Ni nanoparticles calculated with (111) reflection. Red squares represent small NP, and blue squares represent large NPs.



**Figure S6.** Temperature-dependent evolution of mesoporous graphene structures during thermal conversion of  $[\text{Ni}_2(\text{EDTA})]$  to *mesoG*/Ni. TEM images after heat treatment from 400 °C to 1000 °C with 100 °C increments.



**Figure S7.** Durability of electrocatalytic activity for OER. (a) The 1st (black) and 100th (red) CV scans of 3D *mesoG*. (b) The 1st (black) and 100th (red) CV scans of Ir/C catalyst. (c) The 1st (black) and 100th (red) CV scans of Pt/C catalyst.

In the initial OER polarization curve of 3D *mesoG*, a carbon oxidation current was clearly observed at 1.2~1.55 V. However, after 100 subsequent cycles, a redox couple developed around 1.4 V, and the carbon oxidation current disappeared (Figure S7a). This could be attributed to the partial oxidative corrosion of the carbon shell containing Ni NPs, resulting in the exposure of Ni species covered by the carbon shell, and subsequent Ni<sup>II</sup>/Ni<sup>III</sup> redox reactions. The 3D *mesoG* sample exhibited a nearly unchanged OER polarization curve, whereas significantly diminished activities were observed for Pt/C and Ir/C catalysts after 100 potential cycles.

## References for Supporting Information

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