

Supporting Information

A Non-Heat-Source Process for Preparing Graphene Oxide with Low Energy Consumption

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Table S1. Quantitative analysis of rHGO and rNGO

Sample	Component content (at.%)		
	C	O	S, Cl
rHGO	82.74	16.25	1.01
rNGO	81.72	16.75	1.53

The experimental process was the same as NGO, except that after adding potassium permanganate, 50 mL of deionized water was added at the rates of 2.8 mL min⁻¹, 1.33 mL min⁻¹, and 0.72 mL min⁻¹, respectively. The following steps and washing method were identical to those used in the NGO. The products after washing were freeze-dried for 48 h to obtain NGO-2.8, NGO-1.33, and NGO-0.72, respectively. With the decrease of water addition rate, GO gradually changes from dark green to yellow, indicating that the water addition rate has a significant impact on the composition or structure of GO (Fig. S1.).

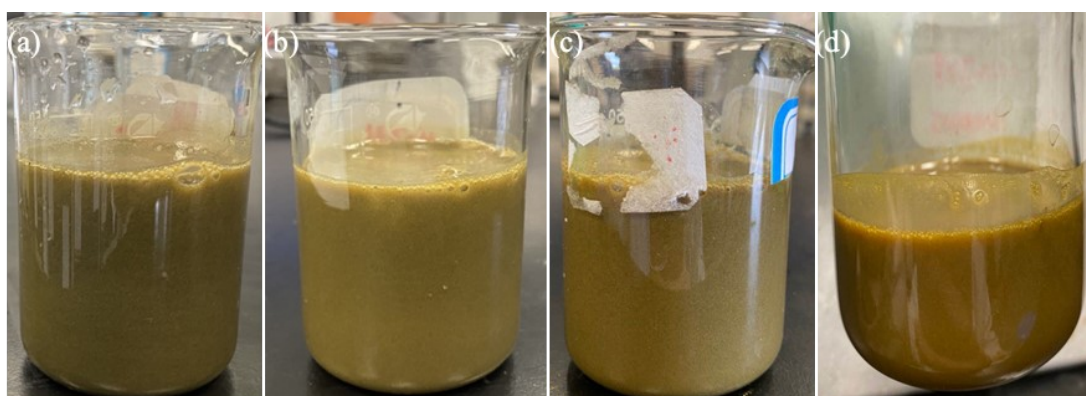


Fig. S1 Optical images of NGO-2.8 (a), NGO-1.33 (b), NGO-0.72 (c) and NGO (d).

As shown in Fig. S2a, NGO-2.8, NGO-1.33, and NGO-0.72 all have a sharp peak at $2\theta = 9\sim 10^\circ$, indicating that they are GO. However, the diffraction peaks of NGO-2.8,

NGO-1.33, and NGO-0.72 is the result of the superposition of the two peaks. Obviously, there is a slight bump at about $2\theta = 10^\circ$, indicating that the oxidation is not uniform in NGO-2.8, NGO-1.33, and NGO-0.72 and partial stacking occurs, leading to diffraction peaks of different degrees. In addition, NGO-2.8 has a weak peak at $2\theta = 26.3^\circ$, which corresponds to the (002) crystal plane of graphite, indicating that it contains a part of unoxidized flake graphite.¹⁻² This suggests that adding water rate too fast will lead to incomplete reaction and graphite cannot be homogeneously oxidized. As shown in Fig. S2b, the calculated I_D/I_G values of NGO-2.8, NGO-1.33, NGO-0.72 and NGO were 0.95, 0.98, 1.01, and 1.03, respectively. Thus, with the decrease of water addition rate, the defect degree gradually increases, which indicates that the oxidation degree increases, resulting in more defects and disordered structures.

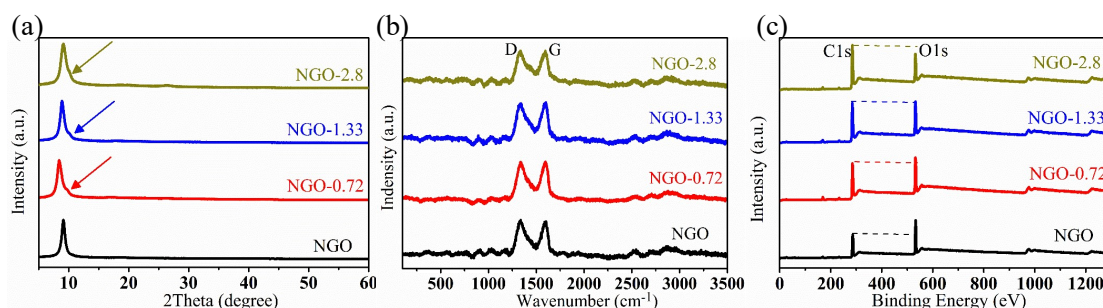


Fig. S2. (a) XRD patterns of NGO-2.8, NGO-1.33, NGO-0.72, and NGO. (b) Raman spectra of NGO-2.8, NGO-1.33, NGO-0.72, and NGO. (c) XPS surveys of NGO-2.8, NGO-1.33, NGO-0.72, and NGO

The main components of the four are C and O, but obviously, with the increase in water addition rate, the peak intensity of C1s is close to that of O1s, even surpassing O1s in the XPS spectrum of NGO-2.8 (Fig. S2c). Through quantitative analysis, the C/O ratios of NGO-2.8, NGO-1.33, NGO-0.72, and NGO are 4.00, 3.25, 2.80, and 2.18,

respectively, which indicates that their oxidation degree increases successively with the slowing down of the water addition rate.

Through SEM observation, the morphologies of the four differ greatly. There are a lot of folds in NGO-2.8, but there is no clear separation between layers. This rate cannot produce enough oxygen containing functional groups to overcome the van der Waals force between the layers. Thus, NGO-0.72 and NGO-1.33 have begun to separate from the accumulated matrix to form a separate layer. When the water rate is 0.28 mL min^{-1} , the GO can be fully expanded and completely separated into lamellae. Therefore, the rate of adding water to prepare GO by this method must be less than or equal to 0.28 mL min^{-1} .

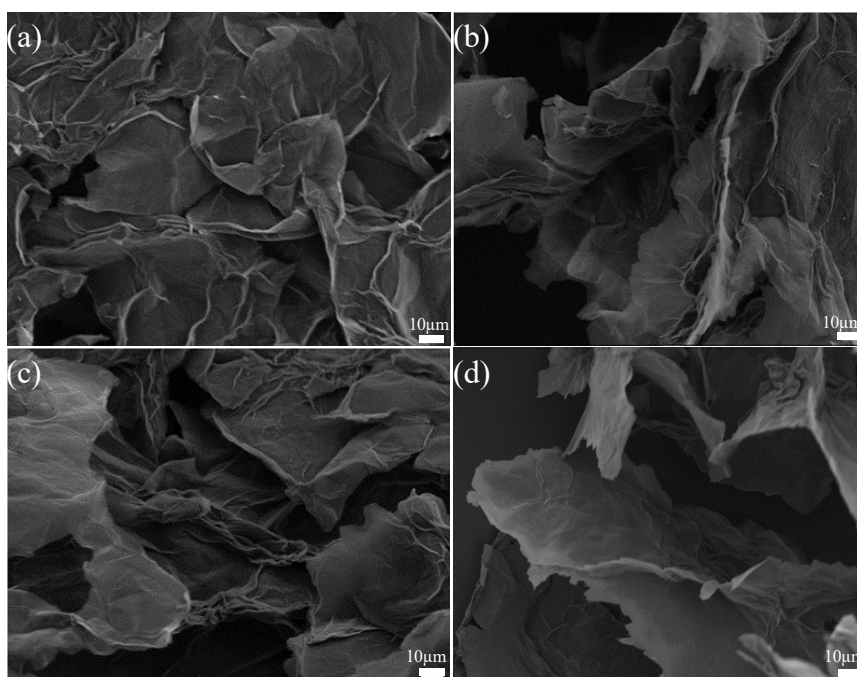


Fig. S3 FE-SEM images of NO-2.8 (a), NO-1.33 (b), NO-0.72 (c), and NGO (d).

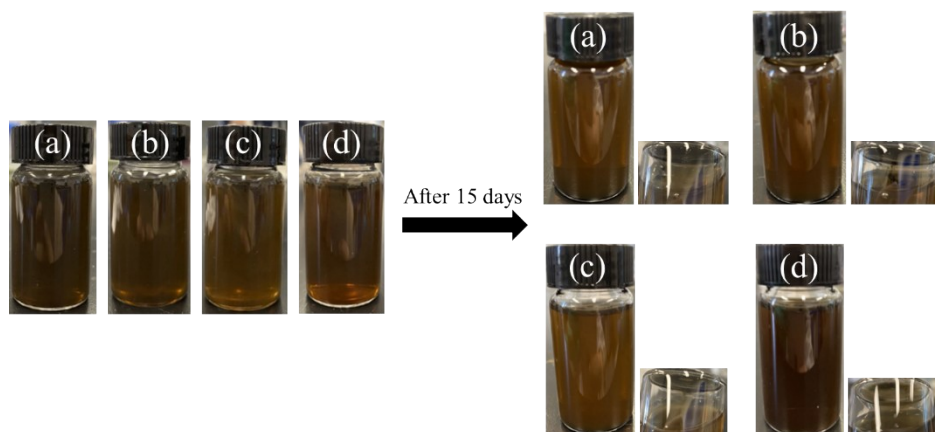


Fig. S4 The dispersion of NGO-2.8 (a), NGO-1.33 (b), NGO-0.72 (c), and NGO (d).

As shown in Fig. S4, four GOs were dispersed into deionized water at a concentration of 100 mg mL^{-1} , respectively. It can be found that they are uniformly permeable when they were just dispersed into water. However, after 15 days of standing, precipitation was observed at the bottom of each sample, but in different amounts. NGO-2.8 has the most precipitation, so the color of its solution becomes lighter. This indicates that four GOs have different oxidation states.

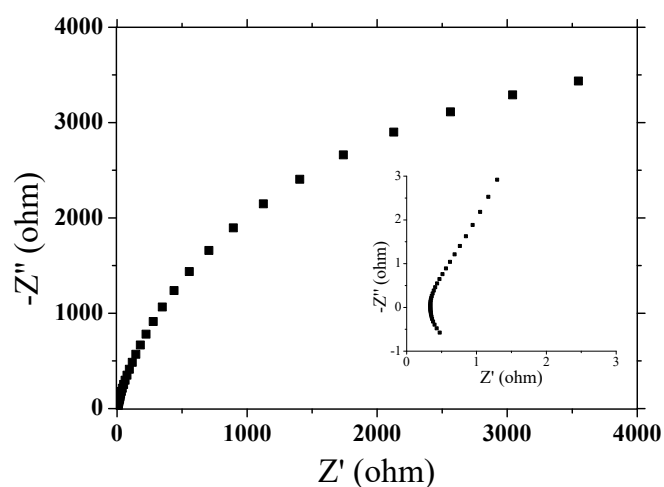


Fig. S5 EIS spectrum of the naked Ni foam.

References

1. Zaaba, N.; Foo, K.; Hashim, U.; Tan, S.; Liu, W.-W.; Voon, C., Synthesis of graphene oxide using modified hummers method: solvent influence. *Procedia Engineering* **2017**, *184*, 469-477.
2. Al-Gaashani, R.; Najjar, A.; Zakaria, Y.; Mansour, S.; Atieh, M., XPS and structural studies of high quality graphene oxide and reduced graphene oxide prepared by different chemical oxidation methods. *Ceramics International* **2019**, *45* (11), 14439-14448.