Supplementary Information

Chemical reduction of an aqueous suspension of graphene oxide by nascent hydrogen

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Descriptions of graphene oxide synthesis, Photograph of reduction of graphene oxide by nascent hydrogen, SEM images of $nHRGO_{Al/NaOH}$ before and after washing with diluted HCl solution and XRD patterns of nHRGOs.

1. Preparation of graphene oxide

A small amount of expandable graphite was charged into a 1-L beaker and heated for 10 s in a microwave oven. The graphite expanded to about 150 times its original volume. Graphene oxide was synthesized from expanded graphite according to a modified Hummers method .[S1] Typically, 1 kg of concentrated H_2SO_4 (95 wt.%) was charged into a 2L beaker equipped with a mechanical stirrer (Teflon impeller). The beaker was put into an ice bath to chill to 0°C. Seven grams of expanded graphite were gradually added under stirring to make a suspension. Then, 45 g of $KMnO_4$ was slowly added so that the temperature did not exceed 20°C. The temperature was then elevated to 35°C, and the suspension was stirred for 2 h. The beaker

was then chilled again in the ice bath, and 1.5 L of deionized water was slowly added to maintain a temperature below 70°C. The mixture was stirred for 1 h and subsequently diluted with 10 L of deionized water. Fifty milliliters of H_2O_2 (30 wt%) was slowly added, and vigorous bubbles appeared as the color of the suspension changed from dark brown to yellow. The suspension was centrifuged and washed with 1M HCl solution four times, followed by centrifuging at 10,000 rpm and washings with deionized water to completely remove the acid until the pH of the GO dispersion reached 6. The as-synthesized GO dispersion was a paste. The concentration of GO was approximately 1.0 wt%, which was determined after drying the GO dispersion at 80 °C under vacuum for 24 h.

2. Reduction of graphene oxide by nascent hydrogen

Fig. S1(a)&(b) show that Al foils highly reflected the camera's flash light, indicating the surface of Al foil were very clean, not covered by graphene oxide sheets. Similarly, the enlarge of Fig. S1(c) shows that there are several bright dots which are Zn particles reflected camera's flash light, implying the Zn particles were not wrapped by graphene oxide sheets. Since there is no adhesion of graphene oxide sheets on the Al and Zn surfaces, the reduction of graphene oxide by electron transport directly from Al and Zn to graphene oxide sheets as reported in previous literatures [S2-S4] can be excluded.

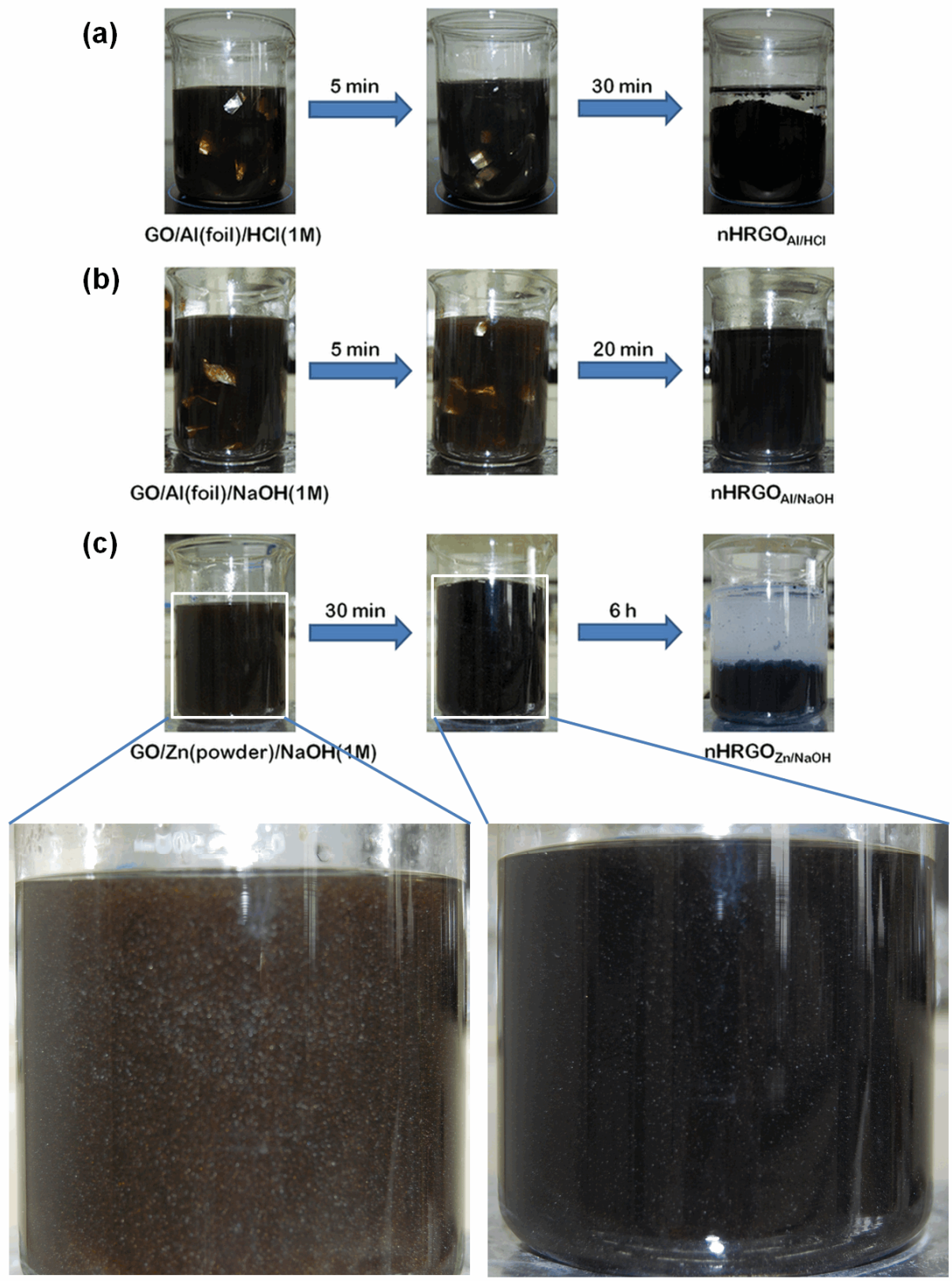

Fig. S1. Photographs of the graphene oxide reduction processes by nascent hydrogen

3. Washing residual salt by diluted HCl solution

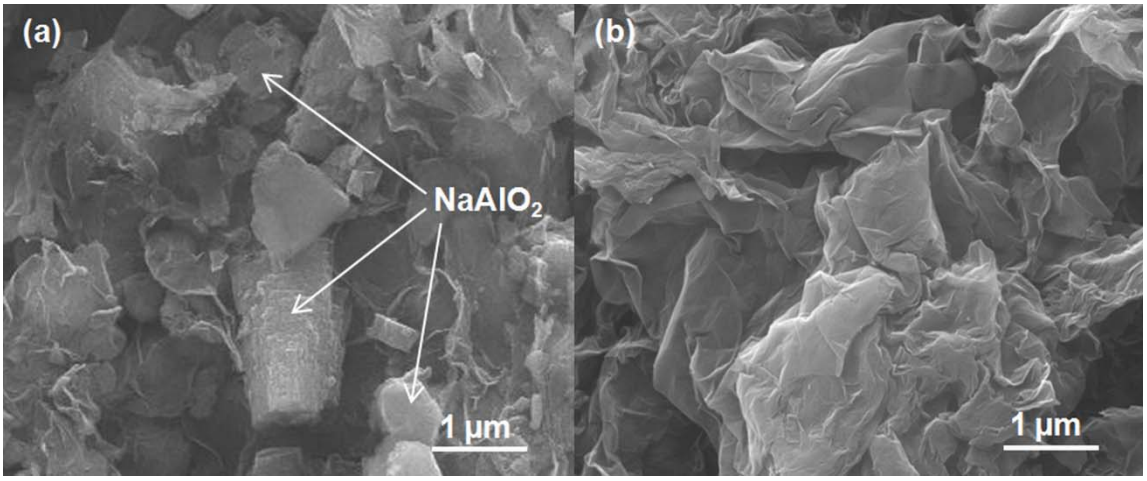

Fig. S2. The morphology of $nHRGO_{Al/NaOH}$ before (a) and after (b) washing three times with 0.1 M HCl solution

The reactions between Al with NaOH and Zn with NaOH produced $NaAlO_2$ and Na_2ZnO_2, respectively (Scheme 1). These salts have low solubility in alkaline condition, so they precipitated into particles and attached on nHRGO sheets. Attempt to washing with DI water could not remove these salts (Fig. S2(a)). However, these salts are easily removed by washing with diluted HCl solution (Fig. S2(b)).

4. XRD spectra of nHRGOs

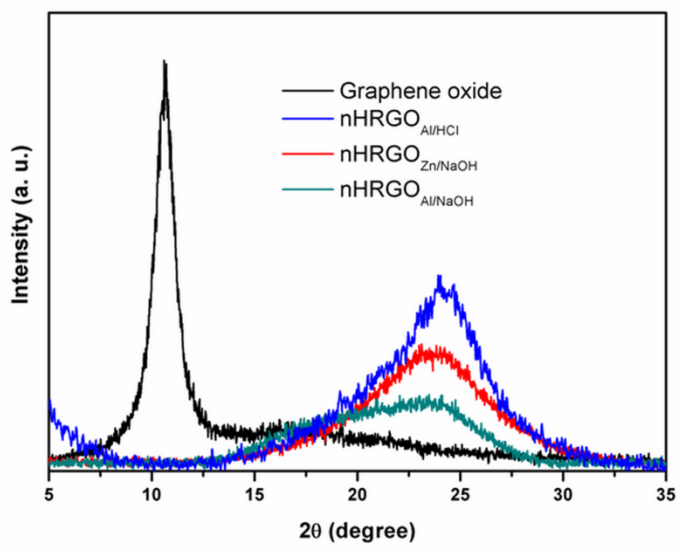

Fig. S3. XRD patterns of nHRGOs

The XRD spectrum of graphene oxide exhibits an intense peak at 11.0^{o}, corresponding to d-spacing of 8.07A due to the interlamellar water trapped between the hydrophilic graphene oxide sheets. In the XRD spectra of nHRGOs, the $d_{spacing}$ peak were broad and dramatic shifted to higher 2θ angle, at around 24^{o}, close to the dspacing peak value of graphite (26.7^{o}),[S5] suggesting that nHRGOs were highly graphitized. The intensities of dspacing peaks of nHRGOs reveals that the graphitization degree of nHRGOs are in order of nHRGOAl/HCl > nHRGOZn/NaOH > nHRGOAl/NaOH which is consistent with the elemental analysis, XPS, Raman, TGA and electrical conductivity results.

References

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[S2] Fan Z, Wang K, Wei T, Yan J, Song L, Shao B. An environmentally friendly and efficient route for the reduction of graphene oxide by aluminum powder. Carbon 2010; 48(5):1686-89.

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