

Supporting information for

Ion Diffusion-Directed Assembly Approach to Ultrafast Coating of Graphene Oxide Thick Multilayers

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1. Relationship between slop of pulling force-distance curve and the gel circumference

The substrate, a copper wire, was pre-treated: it was dipped into GO dispersion and then pulled out, to adhere a quite thin GO layer. After adsorbing electrolyte solution, the copper wire was fixed on the up clamp with a force sensor of the testing machine. The GO dispersion bath was fixed below. The wire quickly dipped 30 mm inside the coating bath at a speed of 600 mm min⁻¹. Thus the “dipping in” process costs 3 s. The time from when the dipping in finishes to when pulling-out starts is termed as detention time. The pulling-up speed is 60 mm min⁻¹. Thus indeed, the immersion time of the tail of the copper wire is 30 s (30mm / (60 mm min⁻¹)) longer than that of the head.

The pulling distance is denoted as D . The measured dragging force (F) is a sum of the gravity of the gel (mg) lifted out of the GO dispersion bath surface and the shear force between the GO gel immersed in bath and GO dispersion.(see Equation S1) Under a constant pulling velocity, the shear stress (τ) can be deemed as a constant value. Thus the shear force is proportional to the area (A) of moving interface between gel and GO dispersion. Its value gradually decays during pulling-out process (Figure 2g). Note that the value of the gravity part (0-0.3 mN, measured by an electronic balance) is negligible compared with the shear force (0-8 mN) in this case. Thus, the slope (*i.e.*, $\frac{dF}{dD}$) of the force-distance curve is proportional to the circumference (l) of gel shell leaving the bath surface, as expressed by Equation S2.

$$F = mg + A \times \tau \approx A \times \tau \quad (1)$$

$$-\frac{dF}{dD} = l \times \tau \quad (2)$$

When the detention time is ≤ 25 s, the tail has a thicker coating than the head; When the detention time is ≥ 30 s, the tail shares the same diameter as the head. Correspondingly, the force-distance curve holds a constant slope when the detention time is ≥ 30 s, indicating l does not change during the pulling-out process.

2. Relationship between *sdc* and critical coagulation concentration (*ccc*).

The coagulation concentration is measured as follows. The electrolyte was added into GO dispersion with GO concentration of 0.05 mg mL^{-1} . After being mixed by a vortex mixer and settled for 2 minutes, the dispersion was slightly centrifuged to accelerate the settlement of possible aggregation. The concentration of electrolyte when aggregates emerge was deemed to be the *ccc*. The results are listed in Table 1.

To understand the relationship between *sdc* and applied electrolyte concentration in Figure 3a, we propose a formula as follows.

$$G_2 = G_1 \times f \times \frac{C}{sdc} + A$$

In which, G_1 is the weight of the adsorbed electrolyte solution in the GO-film pre-coated copper wire, and measured to be 0.046 mg, G_2 is the coated gel mass, f is the fraction of ions participating in diffusion/gelation of whole adsorbed ions (considering that there are ions captured in the reservoir when IDDA finishes), C is concentration of the applied electrolyte solution, and A is the mass of the adhesive

layer outside the ion-crosslinked gel coating. Note that A can be treated as a constant, ignoring a slight change of the gel shell diameter.

The value of slope ($=G_1 \times f / sdc$) is obtained through a linear fitting of the curves in Figure 3a. As in the case of GO film-coated copper wire, f can be deemed to be constant for different ions of BaCl_2 , CaCl_2 and CuSO_4 . When f is taken as 0.5, the calculated sdc (see Table S1) is close to their ccc . Thus, we hypothesis that the nature of sdc may be the same as ccc .

Table S1. Critical coagulation concentration and estimated sdc of electrolytes.

Species	ccc (mM)	slope	sdc (mM, $f=0.5$)
NaCl	670	---	
NH_4Cl	200	---	
CuSO_4	7.2	0.00408	5.60
CaCl_2	4	0.00465	4.60
BaCl_2	1.67	0.01505	1.52

3. Electrochemical measurement of reduced GO (RGO) supercapacitor

The coated GO gel on microporous film was reduced by 55% aqueous hydroiodic acid under 95 °C for 2h, then sufficiently washed by water and ethanol. The edge of the film was cut off by a knife, to electrically separate the two RGO film. Then the sandwich “RGO-separator-RGO” can be tested in a home-made supercapacitor testing device with Pt plate as the current collector, on CHI600E.

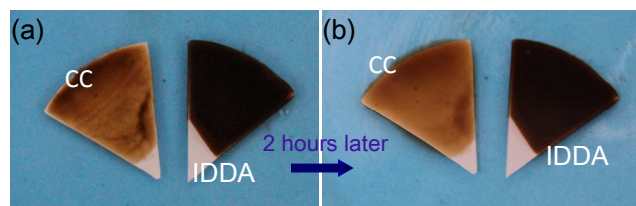


Figure S1. Digital photos of GO gel coating through IDDA and conventional coating (CC) on a cellulose membrane before and after their immersion in water for 2 hours.

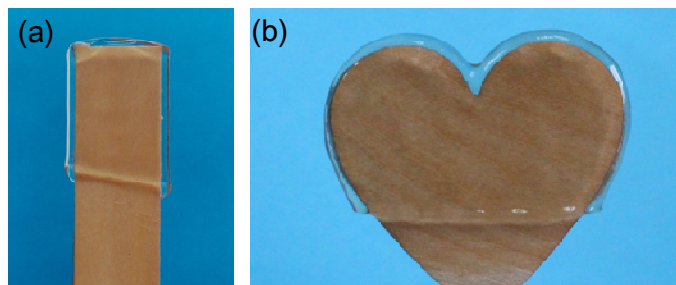


Figure S2. Alginate gel coating produced by IDDA.

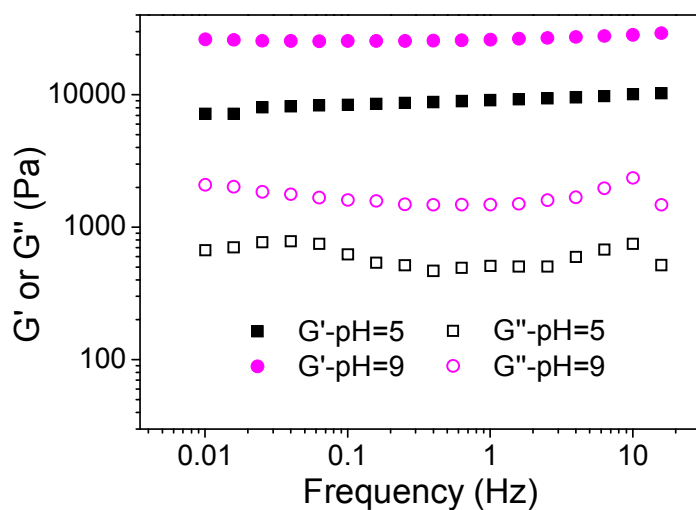


Figure S3. Rheological behavior of Ca^{2+} -crosslinked GO gel produced by IDDA at pH=5 and pH=9.

The rheology properties of the gel were measured by dynamic frequency sweep in the frequency range of 0.01 to 20 Hz at an oscillatory strain of 0.05%. As shown below in Figure 3, the storage modulus (G') of GO gel produced by IDDA at pH=9 is about 25500 Pa, nearly 3 times that at pH=5. The loss modulus (G'') of GO gel produced by IDDA at pH=9 is about 1470 Pa, 2.2-2.8 times that at pH=5.