Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary Information

Selective formation of predominantly pyridinic type nitrogen-doped graphene and its application in lithium-ion battery anodes

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Supplementary Note 1: Experimental setup for high-yield graphene synthesis

In order to fabricate large amounts of GNSPs efficiently, we built a PECVD deposition system with eight chambers in parallel shown in Fig. S1. Input of hydrogen and methane gas is controlled by mass flow controllers (MFCs), and input of 3-chloropyridine is controlled by a leak valve which is connected to a vacuum sealed vial. The pressure is measured by a pressure gauge (PG), and the gas composition is measured by a residual gas analyser (RGA). The gases are split to eight quartz chambers which each have Evenson cavities connected to microwave power sources. A cold trap (CT) captures harmful by products of the reaction (HCl, etc), and a vacuum pump (VP) is continually pumping the system. The pressure is controlled by a throttle valve (TV) that opens and closes with feedback from the pressure gauge to maintain a pressure setpoint. We have also measured the pressure in the quartz chamber, and under the conditions of this experiment (*i.e.*, a pressure of 4.8 Torr at PG and hydrogen and methane flow rates of 48 sccm and 5 sccm, respectively) the pressure in the chamber is ~500 mTorr, consistent with the previous report of Hsu *et al* [14].

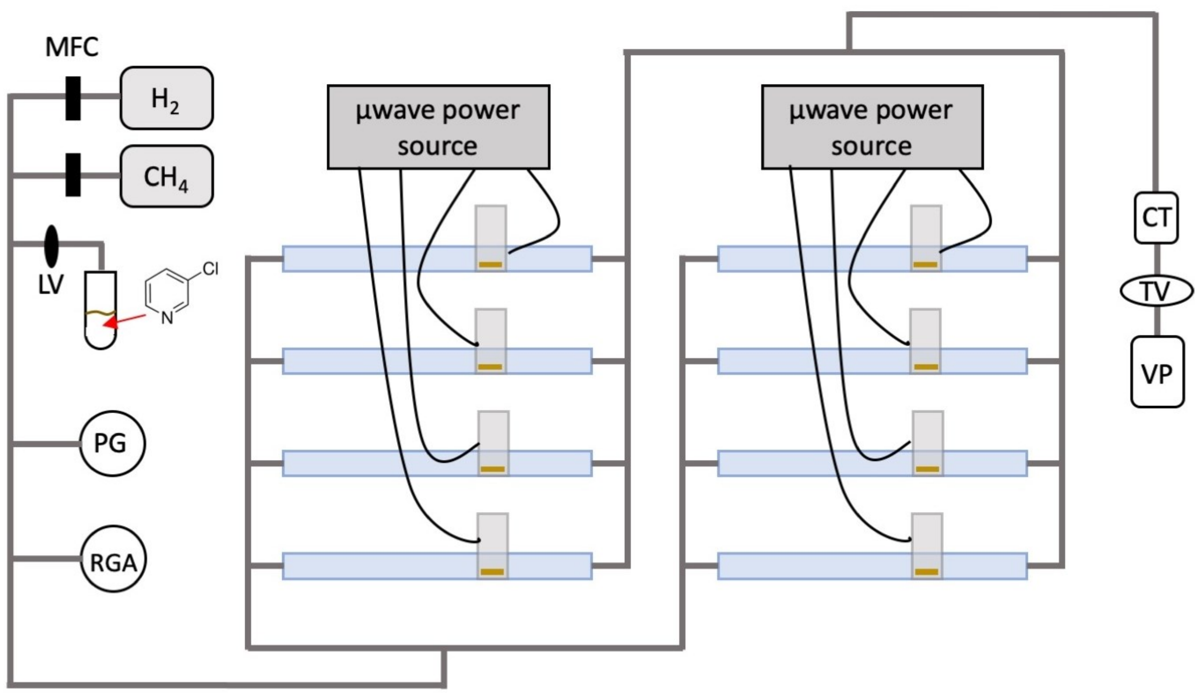

Fig. S1. Schematic of the eight-chamber PECVD graphene growth system.

Supplementary Note 2: Determining the yield of N-GNSPs per chamber

The yield of N-GNSPs synthesized on copper substrates as a function of time is shown in Fig. S2 for one PECVD chamber. A fit of these data reveal a growth rate of ~ 6 $mg/cm^2/hr$. Our deposition chamber affords ~ 1 cm^2 substrates within each plasma cavity, so our eight chamber growth yields a growth rate of ~ 48 mg/hr of N-GNSPs.

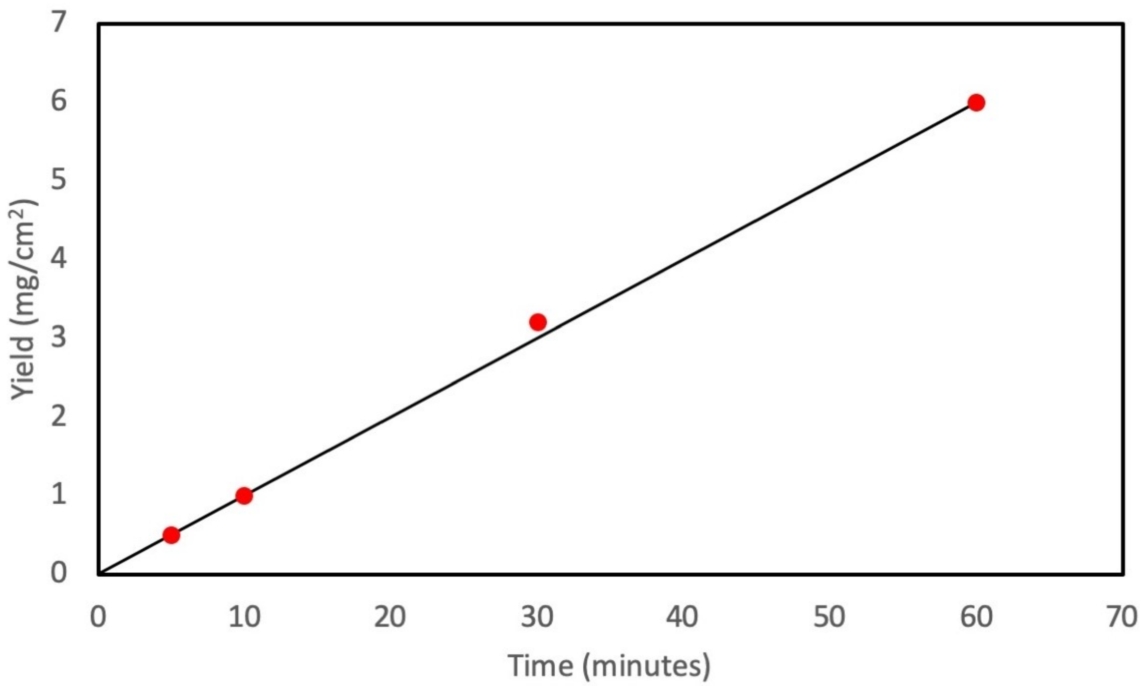

Fig. S2. Yield of N-GNSPs-*vs.*-time obtained from one PECVD growth chamber.

Supplementary Note 3: Procedure for fabricating coin cells of lithium ion batteries (LIBs)

The procedure for fabricating two types of LIB coin cells with N-GNSPs as the anode is schematically shown below in Fig. S3.

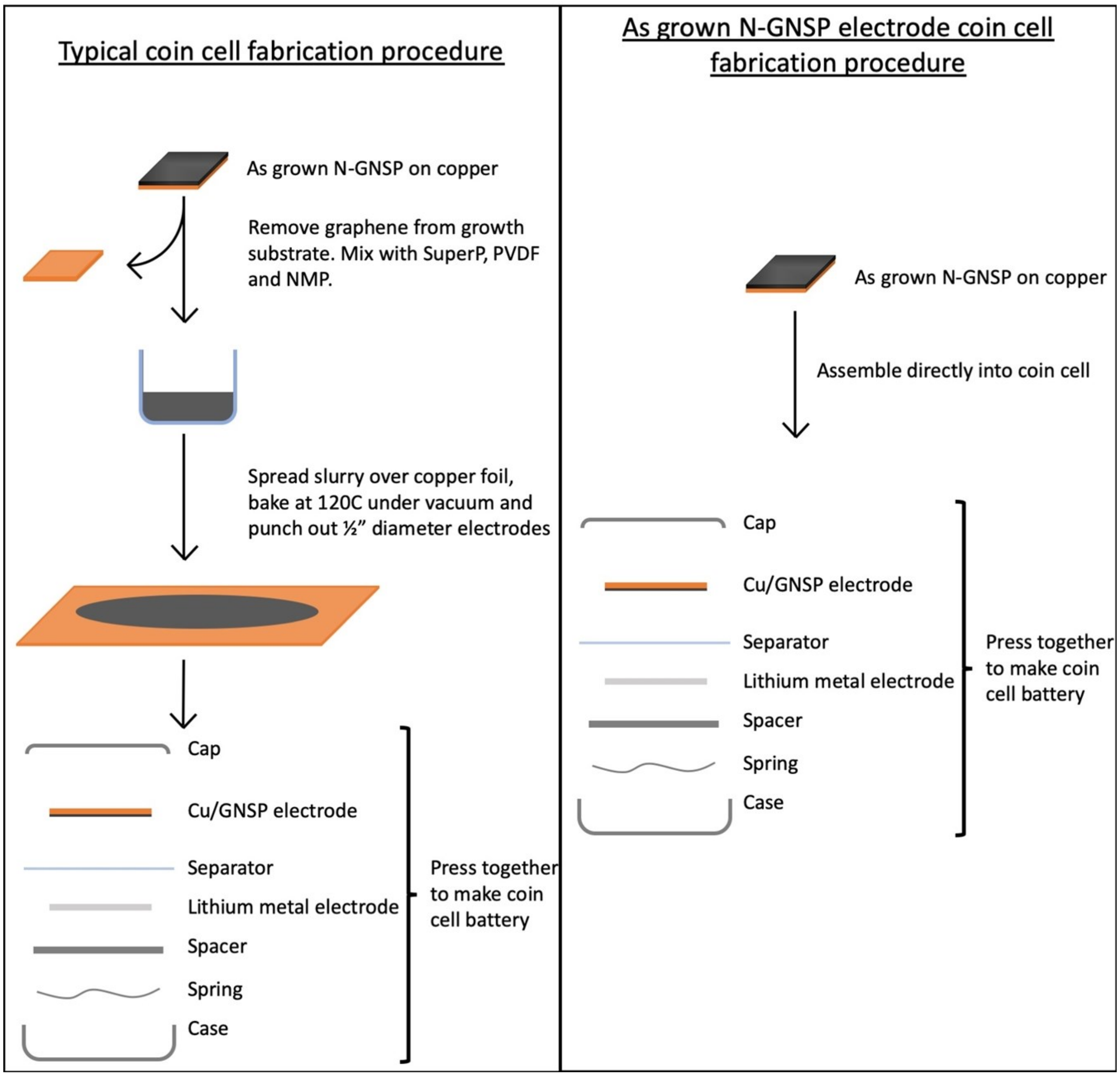

Fig. S3. Schematic of typical coin cell fabrication (left) and fabrication of coin cell using N-GNSP not removed from the growth substrate

Supplementary Note 4: Procedure for estimating GNSPs and N-GNSPs capacitance

According to the widely accepted *Electrochemical Methods* by Bard and Faulkner,[1] double layer capacitance can be estimated from the slope of the voltage *vs*. time response due to a current step. To appropriately perform this measurement, however, the electrochemical response must be due to non-faradaic (capacitive) processes rather than faradaic (redox) processes, which can be ensured by measuring the voltage vs time response shortly after the current step, as non-faradaic processes tend to be much faster than faradaic processes. Therefore, we used the first ten seconds of the first galvanostatic discharge of GNSPs and N-GNSPs to estimate their capacitances (shown in Fig. S4). The fairly linear slopes indicate that the electrochemical response during this time is, in fact, dominated by non-faradaic processes, as faradaic processes tend to cause voltage plateaus in galvanostatic measurements. The capacitance is calculated according to

$$Capactiance = \frac{Current}{Slope}$$

The respective slopes of the GNSPs and N-GNSPs discharge curves are -0.0093 V/s and -0.018 V/s, corresponding to capacitances of 5.6 F/g and 10.8 F/g.

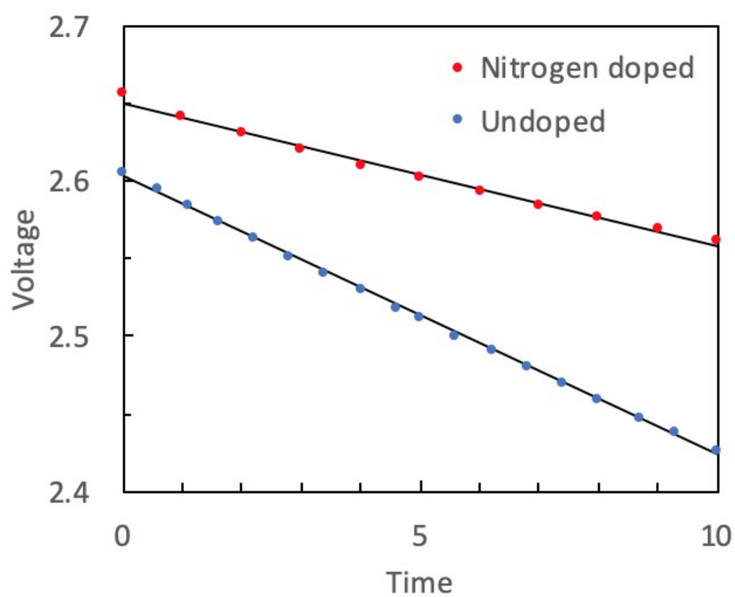

Fig. S4. First ten seconds of GNSPs and N-GNSPs first galvanostatic discharge curve.

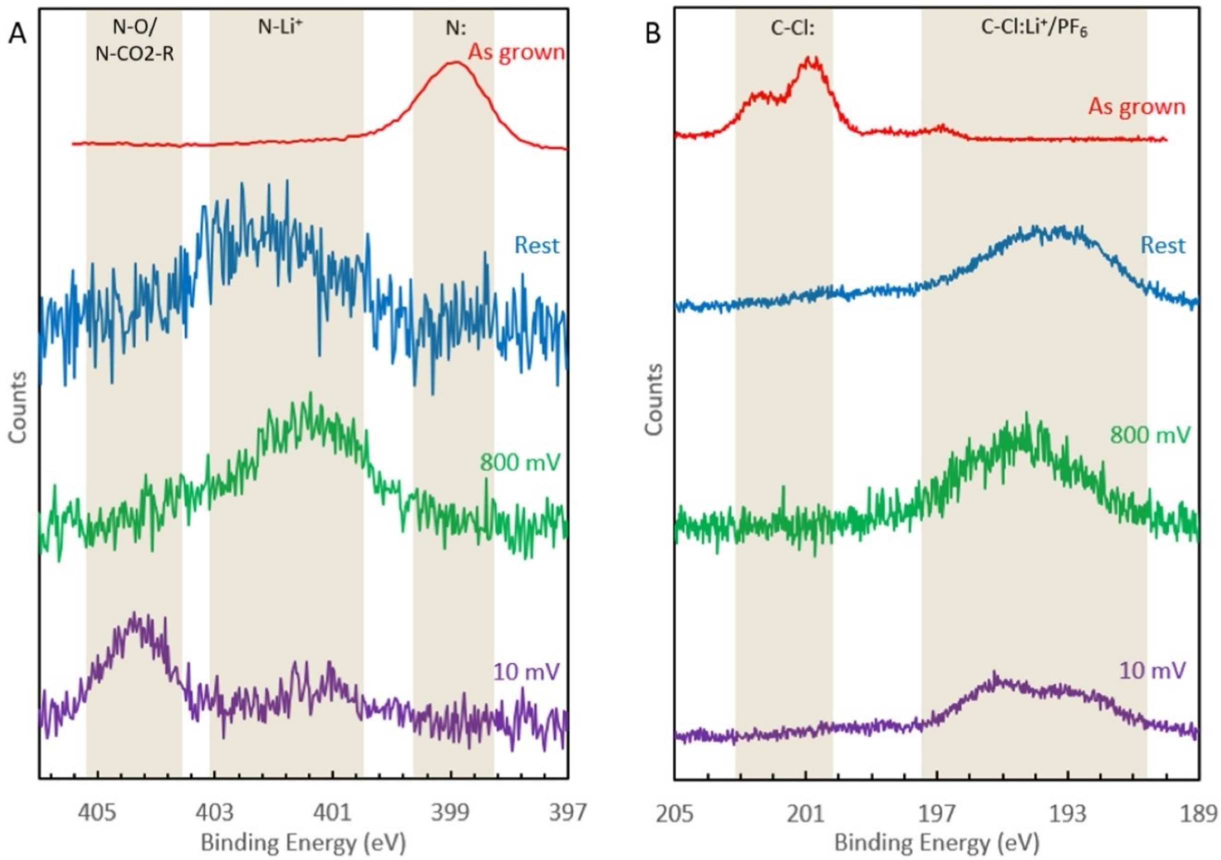

Fig. S5. Raw data (before smoothing) of Fig. 5, showing that the same peaks and shifts are still visible and apparent, which validates the data treatment used to generate Fig. 5.

Reference:

1. *Electrochemical Methods: Fundamentals and Applications*, A. J. Bard and L. R. Faulkner, ISBN: 0471055425, John Wiley & Sons, Inc. (1980).