Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplemental Information for

Sustainable approach for large area transfer of graphene and recycle of the copper substrate

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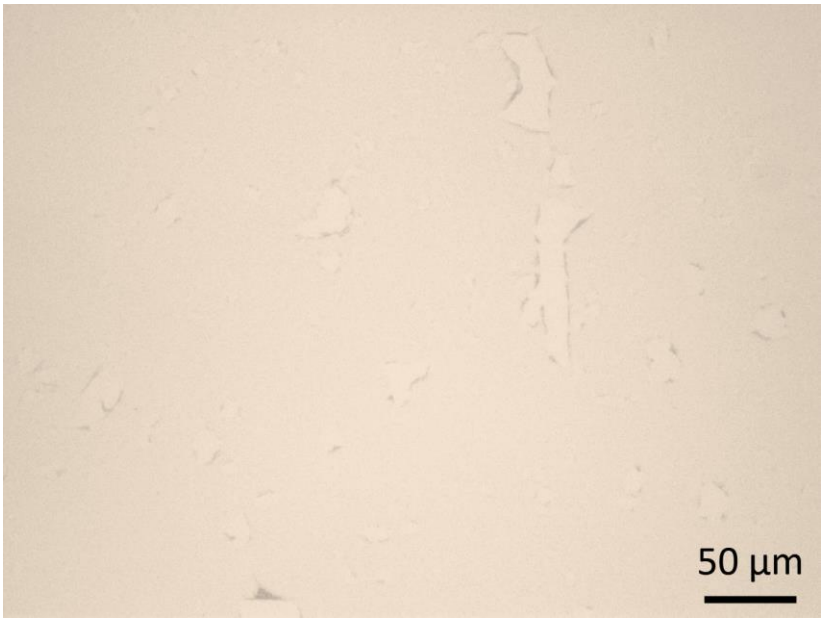

Fig. S1. Optical microscope image of bubbling-damaged graphene with voids transferred onto SiO_2 via bubbling-delamination in 0.1M NaCl electrolyte.

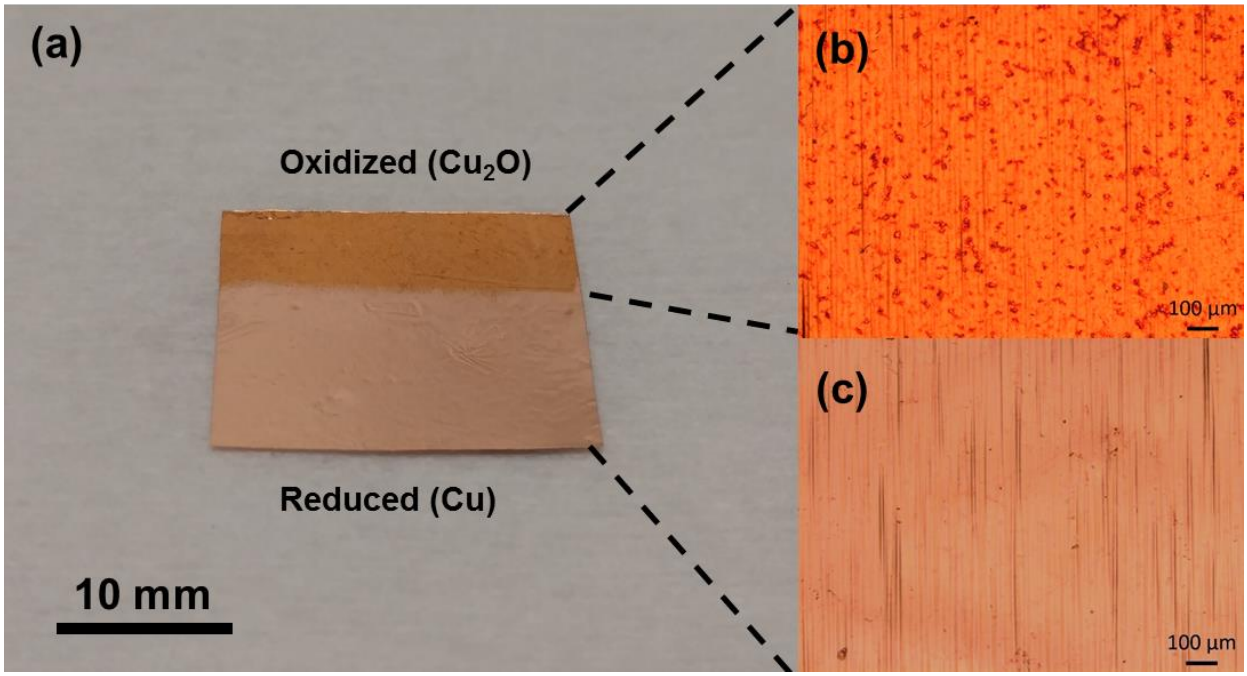

Fig. S2. (a) Partially delaminated ethyl cellulose (EC)/graphene/copper sample showing interface between the (b) oxidized cuprous oxide surfaces (top) where the EC/graphene still remain and (c) the exposed/reduced native copper foil substrate (bottom) where the EC/graphene has been delaminated.

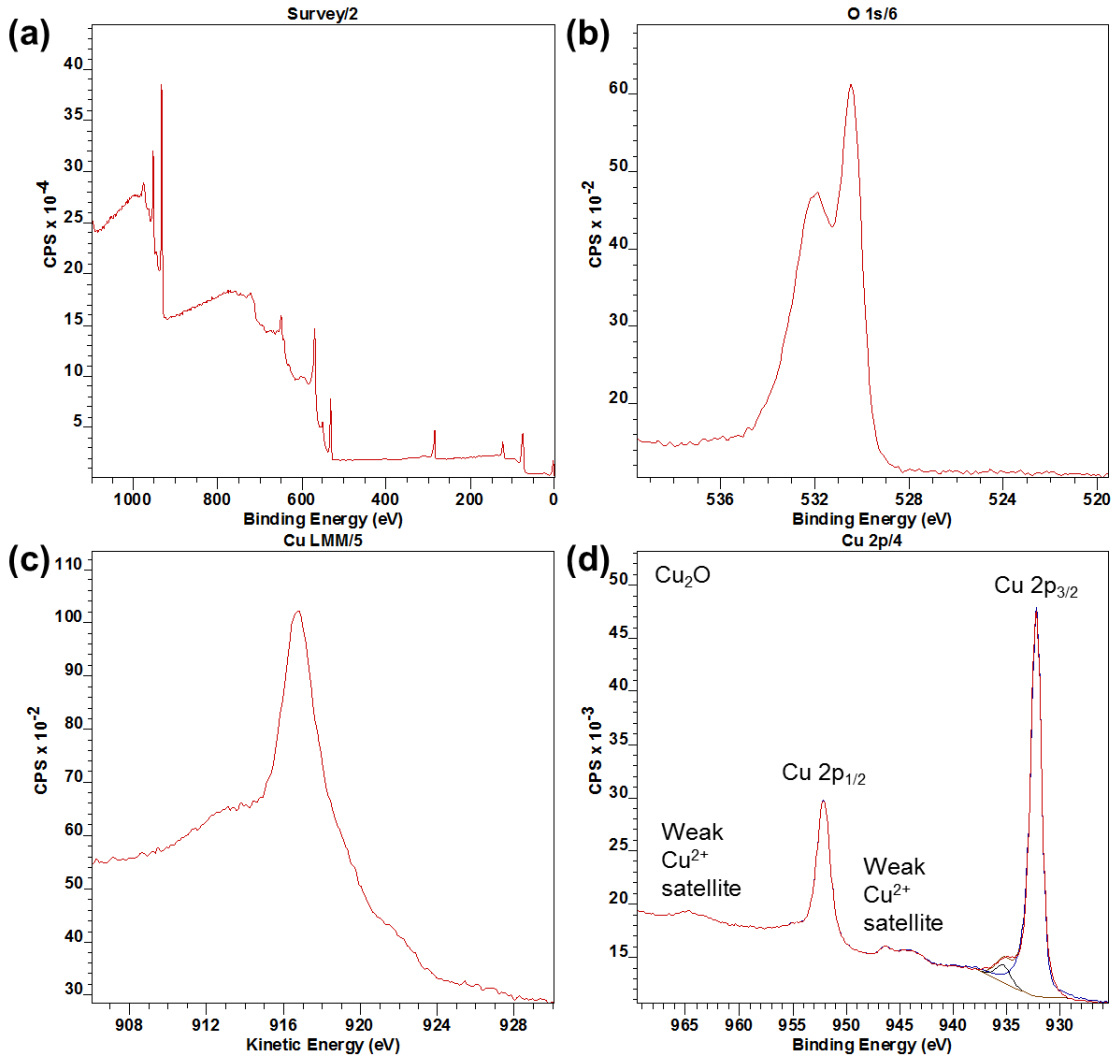

Fig. S3. (a-d) XPS characterization of as-synthesized graphene on copper foil with Cu_2O interlayer due to ambient oxidation. The (c) Cu LMM peak position at less than 917 eV and (d) the weak Cu^{2+} satellites and narrow Cu $2p_{1/2}$ and Cu $2p_{3/2}$ peaks suggest that the oxide layer is predominantly Cu_2O rather than CuO.

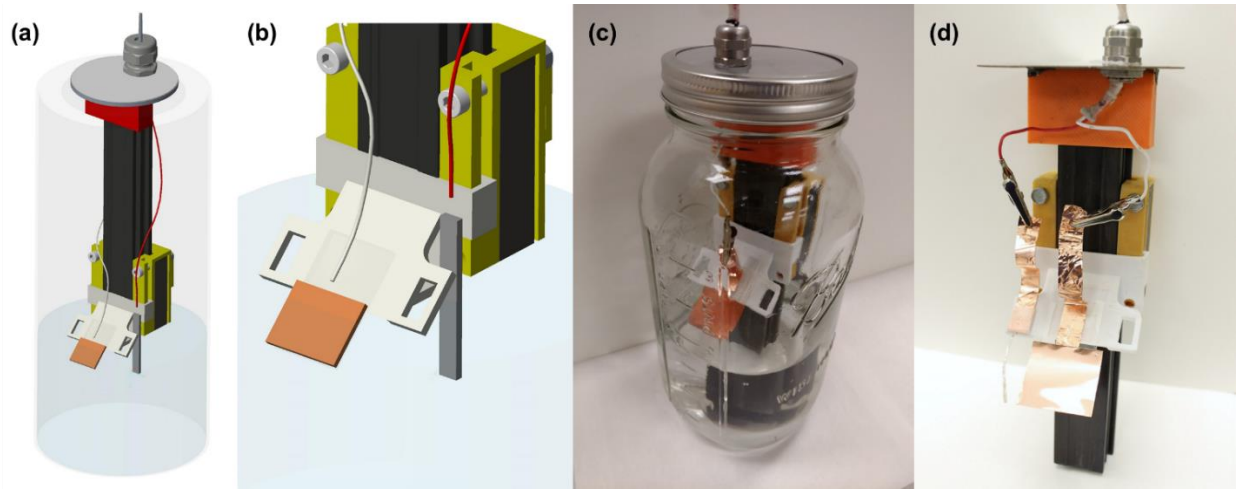

Fig. S4. (a, b) Schematic renderings and (c, d) photos of pressured gradual immersion device for graphene delamination.

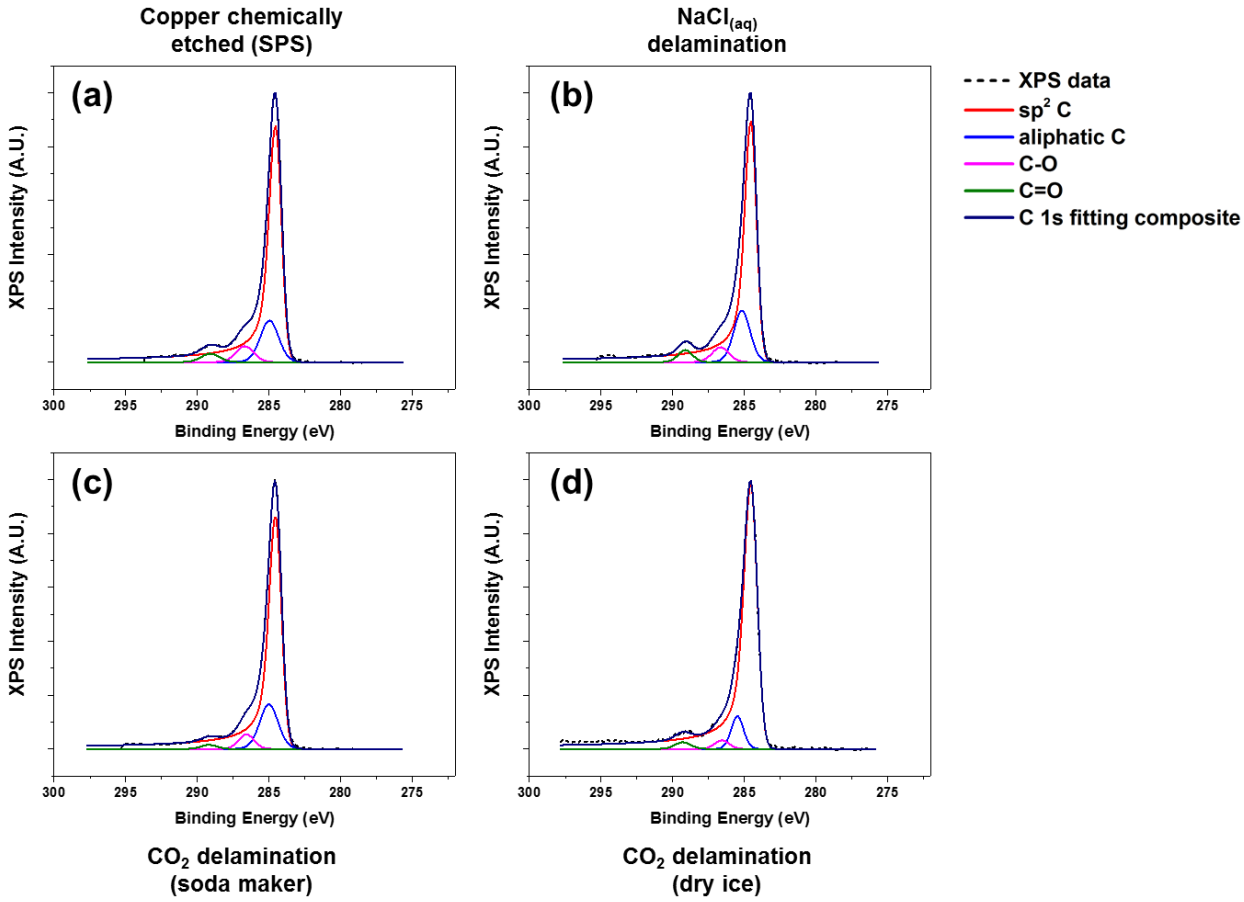

Fig. S5. XPS characterizations of the C 1s peak indicative of graphene and adventitious carbon on the surface of SiO_2 wafer for graphene samples transferred via (a) sodium persulphate chemically etched copper, (b) 0.1M NaCl delamination, (c) CO_2 delamination carbonated with soda maker, and (d) CO_2 delamination carbonated with dry ice.

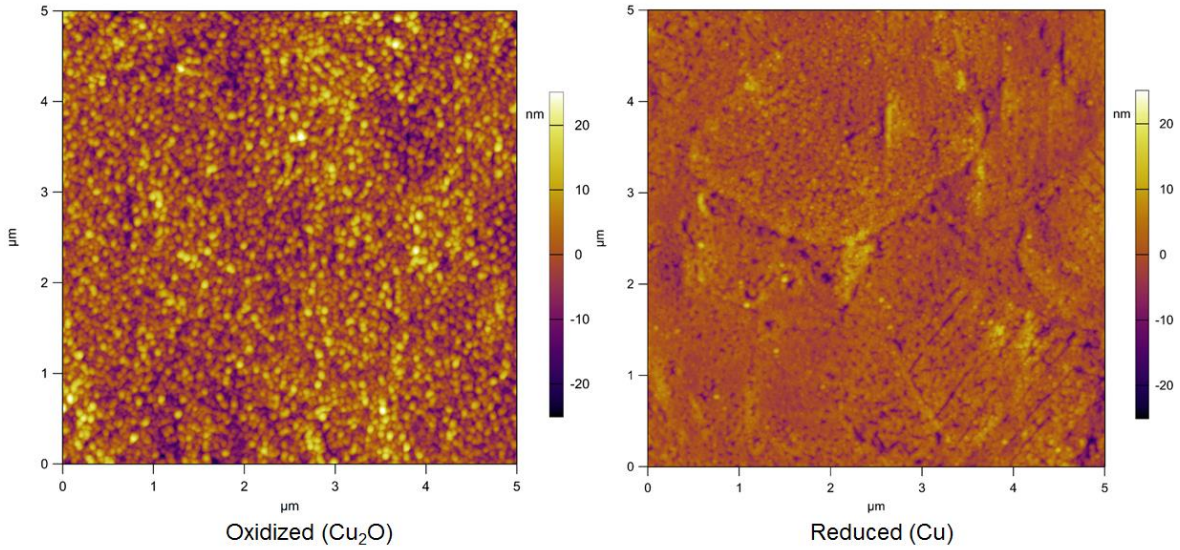

Fig. S6. AFM morphology of oxidized (Cu_2O, left) and reduced (metallic copper, right) copper substrate surfaces, before and after electrochemical reduction in carbonic acid, respectively. Averaged across five random areas of 25um^2 each, the Cu_2O measured a RMS roughness of 6.12±0.03nm whereas the reduced metallic copper surface measured 2.88±0.06nm.

Table S1. Solution conductivities.

Solutions	**Conductivity (μS)**
Fresh DI water	0.1
Fresh DI water + vigorously shaken 20 times	0.7
0.1M NaCl in DI water	9650
DI water + soda maker	72.8
DI water + dry ice	71.9
DI water + dry ice + outgassed 48 hours	33.6
DI water + dry ice + vigorously shaken 20 times	20.9
DI water + dry ice + sonicated 10 minutes	41.2
DI water + dry ice + vigorously shaken 20 times + sonicated 10 minutes	19.8

Table S2. Electrolytes used and applied voltages for various graphene delamination techniques.

Authors	**Electrolyte**	**Applied voltage (cathodic)**	**Reference**
Y. Wang et al.	$K_2S_2O_8$ (0.05mM)	5V	1
L. Gao et al.	NaOH (1M)	5-15V	2
T. Ciuk et al.	NaCl/KCl (2mM-2M)	4-100V	3
X. Wang et al.	Na_2SO_4 (0.5M)	15V	4
C. Cherian et al.	NaCl (0.5M)	2.6V	5
Z. Zhan et al.	NaOH (0.2M)	8V	6
This work	Carbonic acid	7-10V	This work

References

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