

## Monolithic laser scribed graphene scaffold with atomic layer deposited platinum for hydrogen evolution reaction

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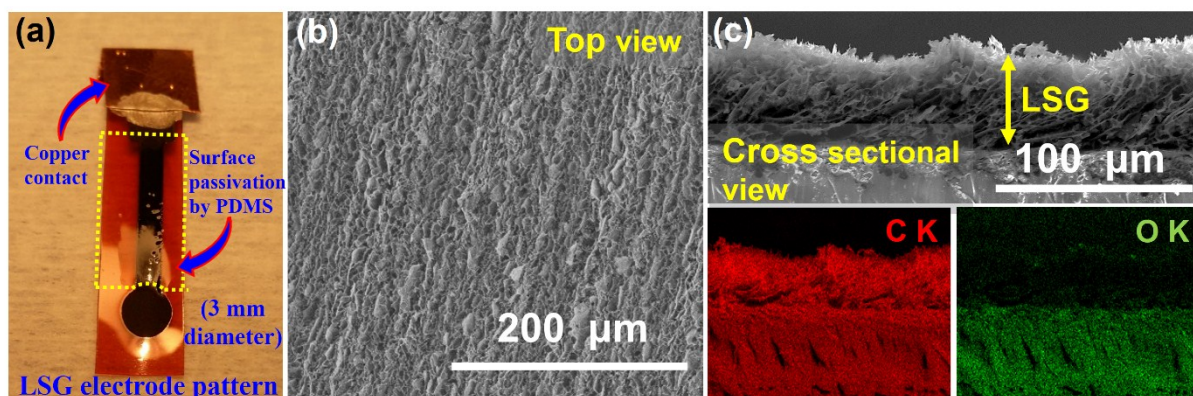
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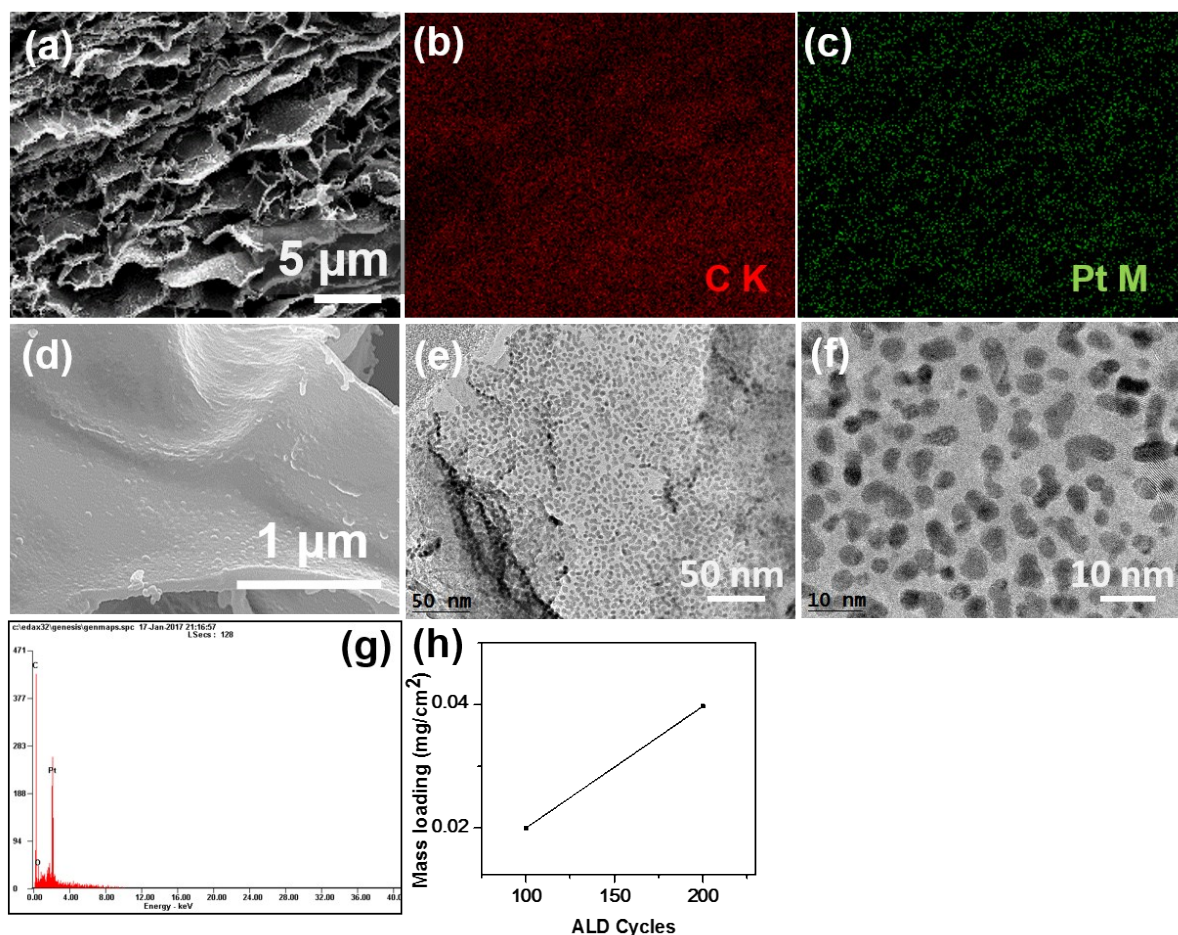
### Electronic Supplementary Information (ESI)



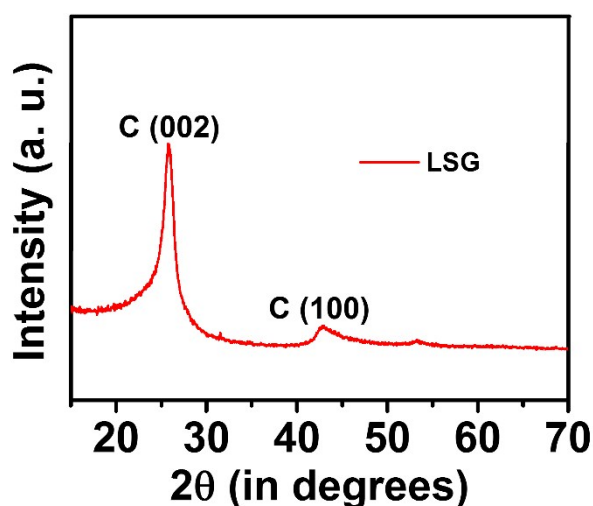
**Fig. S1** (a) Digital photograph of a fully assembled LSG HER electrode scaffold, (b) Top-view SEM micrograph of LSG showing numerous edge plane sites, (c) Cross-sectional view of electrode showing its 3D porous nature and corresponding EDAX maps for C and O elements are shown.

### Preparation of Pt/C@GCE

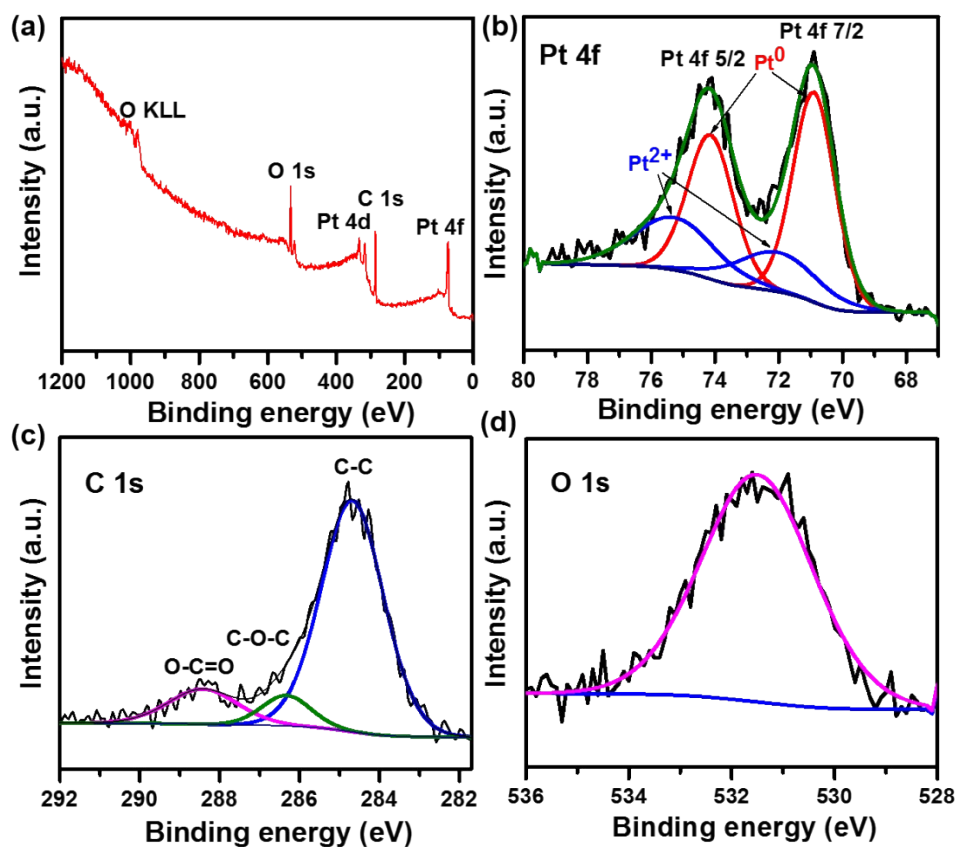
For preparing Pt/C electrode, about 10 mg of commercial Pt/C catalysts were mixed with 0.5 ml of 0.5% Nafion solution. About 10  $\mu$ L aliquot of the dispersion was drop-casted on 3 mm diameter glassy carbon electrode and allowed it to dry under ambient conditions. Prior to use, the Pt/C modified GCE was rinsed in DI water in order to remove the un-attached Pt/C.



**Fig. S2** (a) Top-view SEM micrograph of Pt/LSG<sub>200</sub> electrode showing it is rich in edge plane sites and having ALD Pt on surface, (b) corresponding elemental mapping of C K and (c) Pt M on Pt/LSG<sub>200</sub> surface, (d) high magnification SEM micrograph of Pt/LSG<sub>200</sub> showing surface coated Pt, (e) TEM and (f) HRTEM image of Pt/LSG<sub>200</sub>, (g) EDAX spectrum and (h) plot of mass loading of Pt on LSG after 100 and 200 ALD cycles.



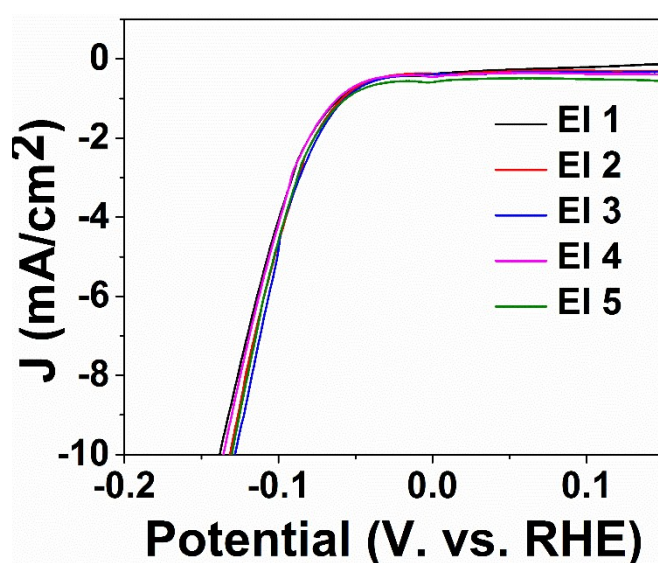
**Fig. S3** XRD pattern of as prepared LSG



**Fig. S4** XPS analysis for Pt/LSG. (a) XPS survey spectra and high resolution XPS spectra for (b) Pt 4f, (c) C 1s and (d) O 1s.

Figure S4 shows the XPS analysis carried out to determine the chemical composition and oxidation states of the elements. As shown in XPS survey scan (Figure S4a), the peaks corresponding to C, O and Pt are present. High resolution XPS spectra can be used to determine

the chemical states of individual elements in Pt/LSG. The deconvoluted spectra of Pt 4f peak (Figure S4b) indicates both Pt<sup>0</sup> and Pt<sup>2+</sup> states. Similarly in deconvoluted C 1s spectrum (Figure S4c), peaks appear at binding energies of 284.6, 286.3 and 288.4 eV corresponding to C=C, C-O-C, O-C=O, respectively. Figure S4d shows the deconvoluted O 1s peak appearing at 531.6 eV.<sup>S1</sup>



**Fig. S5** LSV showing HER onset region for five different Pt/LSG<sub>200</sub> electrodes at 5 mV/s scan rate in 0.5 M H<sub>2</sub>SO<sub>4</sub>.

Table S1. Raman spectra analysis of Pt/LSG.

S. No.	D band (position, FWHM) $\text{cm}^{-1}$	G band (position, FWHM) $\text{cm}^{-1}$	2D band (position, FWHM) $\text{cm}^{-1}$	$I_D/I_G$	$I_{2D}/I_G$
1	1362.0, 41.6	1582.0, 32.1	2722.3, 64.4	0.23	0.35
2	1361.0, 40.1	1579.5, 30.8	2720.1, 57.8	0.21	0.34
3	1361.2, 38.7	1579.6, 30.3	2720.0, 58.3	0.21	0.34
4	1362.2, 37.6	1581.1, 30.0	2722.6, 56.8	0.22	0.35
5	1361.7, 38.2	1580, 29.8	2721.2, 56.2	0.20	0.32

Table S2. Comparison of HER performance (onset potentials) of LSG and Pt/LSG based electrodes.

Electrode	LSG	Pt/LSG <sub>20</sub>	Pt/LSG <sub>50</sub>	Pt/LSG <sub>100</sub>	Pt/LSG <sub>200</sub>	Pt/C
Onset potential (V vs. RHE) at $J = -10\text{mA/cm}^2$	-0.667	-0.188	-0.181	-0.164	-0.131	-0.118

#### References:

S1. D. Longrie, K. Devloo-Casier, D. Deduytsche, S. Van den Berghe, K. Driesen, and C. Detaverniera, *ECS Journal of Solid State Science and Technology*, 2012, **1**, 123.