Electronic Supplementary Information (ESI)

Diffusion controlled multilayer electrocatalysts via sized graphene oxide nanosheets

Minsu Gu,[1]* Jaewon Choi,[1] Taemin Lee,[1] Minju Park,[1] Ik-Soo Shin,[2] Jinkee Hong,[3] Hyun-Wook Lee,[1] and Byeong-Su Kim[1,4]*

[1]Department of Energy Engineering and [4]Department of Chemistry, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Korea

[2]Department of Chemistry, Soongsil University, Seoul 06978, Korea

[3]Department of Chemical Engineering and Biomolecular Engineering, Yonsei University, Seoul 03722, Korea

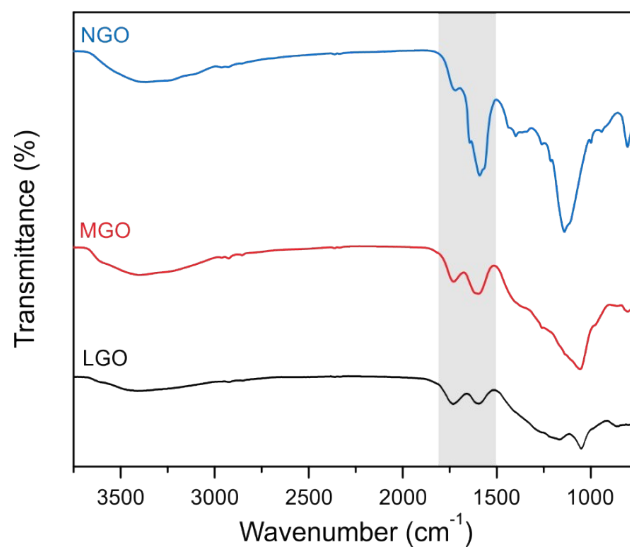E-mail: sbgms@unist.ac.kr (M.G.); bskim19@unist.ac.kr (B.-S.K.)

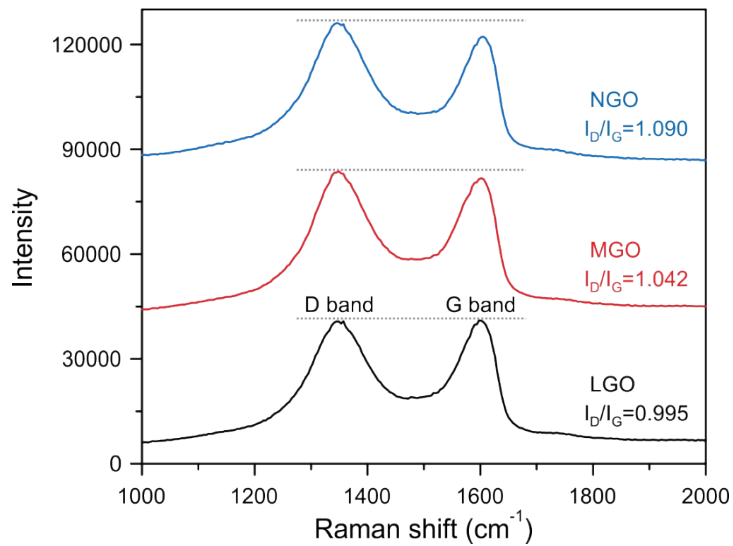**Figure S1.** FT-IR spectra of LGO, MGO, and NGO sheets.

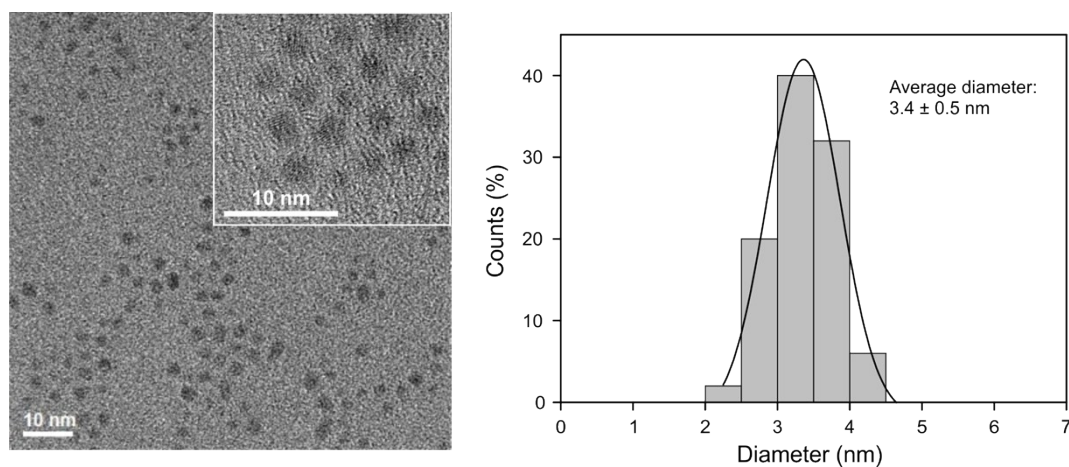**Figure S2.** Raman spectra of LGO, MGO, and NGO sheets.

Figure S3. TEM images with a corresponding size distribution histogram of DMAP-Pd NPs suspension.
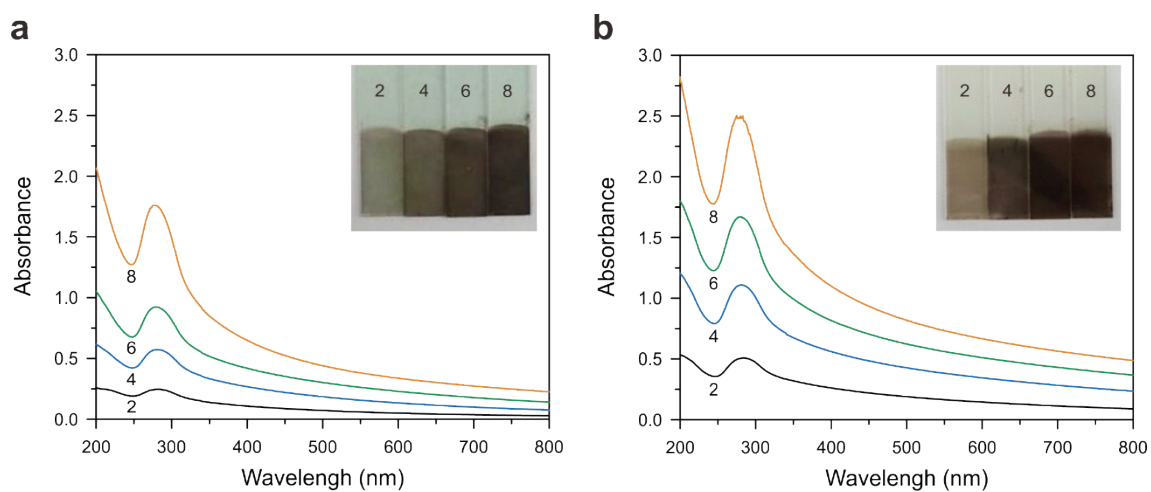

Figure S4. UV/vis absorbance spectra of (a) (MGO/Pd)$_n$ and (b) (NGO/Pd)$_n$ multilayer thin films. Inset image represents the samples prepared.
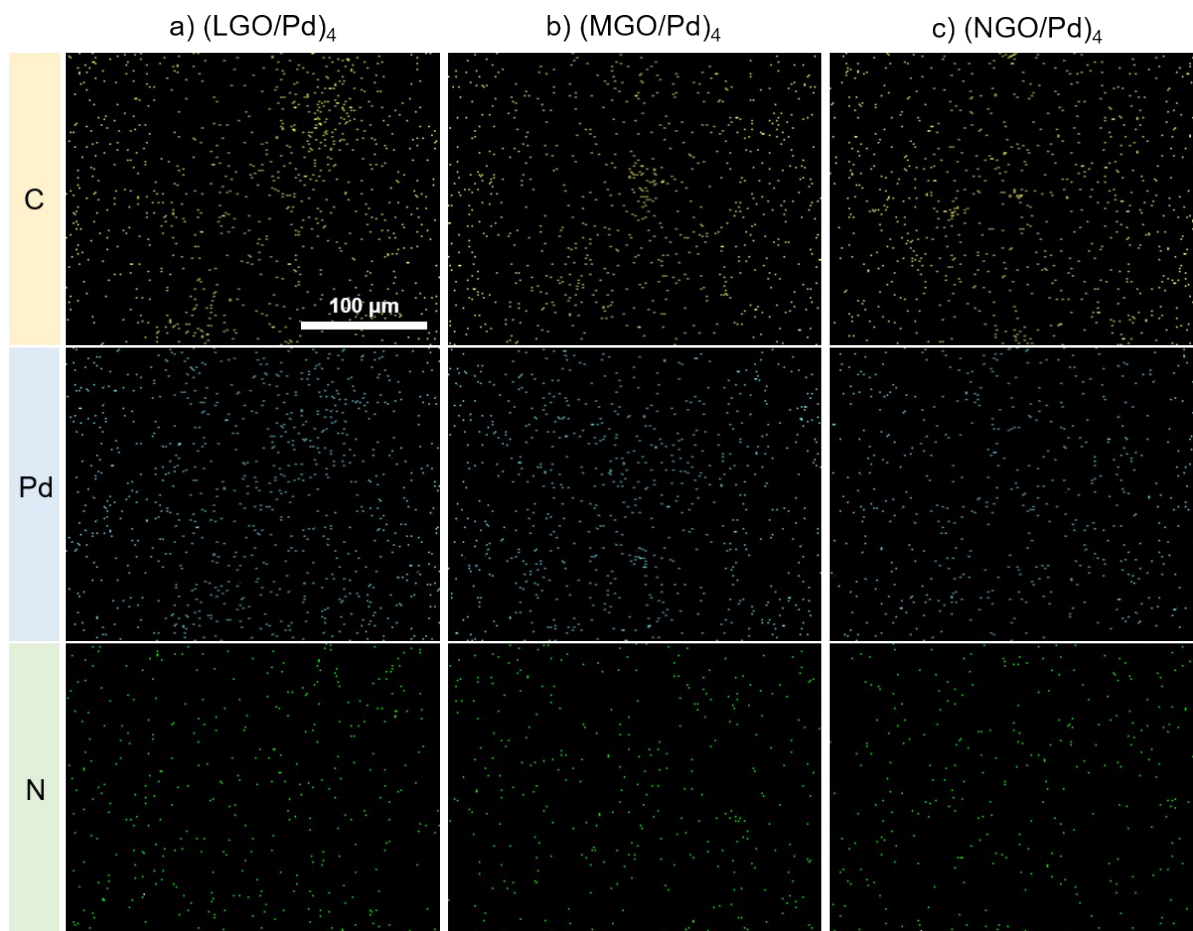
Figure S5. Elemental mapping images of carbon, palladium, and nitrogen of (a) (LGO/Pd)$_4$, (b) (MGO/Pd)$_4$, and (c) (NGO/Pd)$_4$ multilayer thin film electrodes. The scale bar in all images is 100 μm.
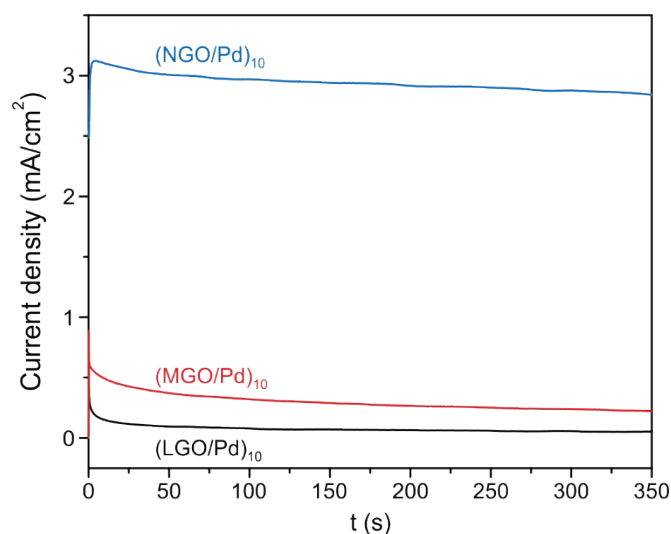
Figure S6. Chronoamperogram (CA) of (LGO/Pd)$_{10}$, (MGO/Pd)$_{10}$ and (NGO/Pd)$_{10}$ multilayer thin films at 0 V (vs. Hg/HgO) in N$_2$ saturated 0.10 M KOH with 1.0 M CH$_3$OH.

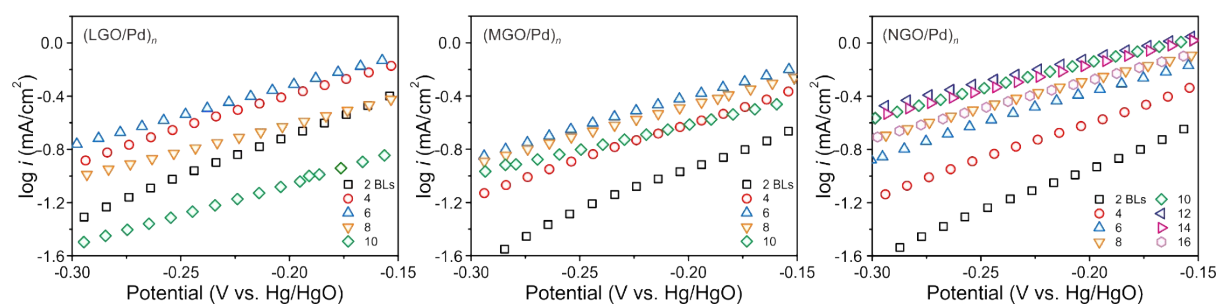**Figure S7.** Tafel plots of (LGO/Pd)$_n$, (MGO/Pd)$_n$, and (NGO/Pd)$_n$ multilayer thin films. Tafel slopes and the corresponding αn were calculated in the Tafel region from -0.30 to -0.15 V.
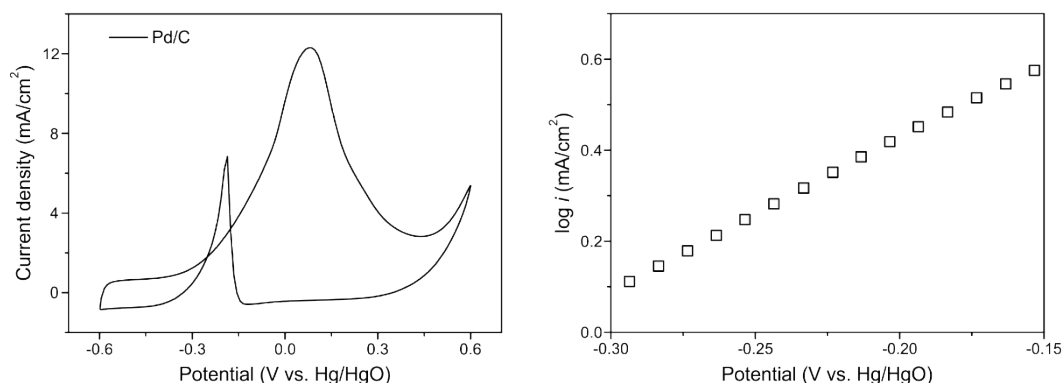
Figure S8. CV and Tafel plots of commercial 30 wt% Pd/C in N_2 saturated 0.10 M KOH with 1.0 M CH_3OH.

Table S1. Properties related to charge and mass-transfer of $(LGO/Pd)_n$, $(MGO/Pd)_n$, and $(NGO/Pd)_n$ multilayer thin films comparing to commercial Pd/C.

Multilayer films	BLs	Tafel slope (mV/dec)	$n\alpha$	Diffusion coefficient (D, m²/s)	Tortuosity, (τ)	Diffusion length (L, nm)
$(LGO/Pd)_n$	6	223	0.264	1.16×10^{-6}	37	1995
	8	249	0.237	1.26×10^{-7}	341	25390
	10	213	0.277	1.47×10^{-8}	2917	241838
$(MGO/Pd)_n$	6	216	0.274	3.44×10^{-6}	12	690
	8	227	0.260	9.66×10^{-7}	44	2837
	10	268	0.220	1.28×10^{-7}	335	24285
$(NGO/Pd)_n$	6	204	0.289	4.48×10^{-6}	10	538
	8	233	0.254	9.09×10^{-6}	5	332
	10	244	0.242	2.07×10^{-5}	2	167
	12	270	0.219	2.43×10^{-5}	2	160
	14	255	0.231	1.21×10^{-5}	4	357
	16	232	0.255	6.40×10^{-6}	7	742
Pd/C		299	0.197	4.29×10^{-5}	1	