Supporting Information for *Acta Phys. -Chim. Sin.* doi: 10.3866/PKU.WHXB202008031

PdNi/石墨烯气凝胶电催化甲酸氧化

包玉菲，冯立纲*

扬州大学化学化工学院，江苏 扬州 225002

Formic Acid Electro-oxidation Catalyzed by PdNi/Graphene Aerogel

Yufei Bao, Ligang Feng *

School of Chemistry and Chemical Engineering, Yangzhou University, Yangzhou 225002, Jiangsu Province, China.

*Corresponding author. Email: ligang.feng@yzu.edu.cn; Tel.: +86-17306299692.

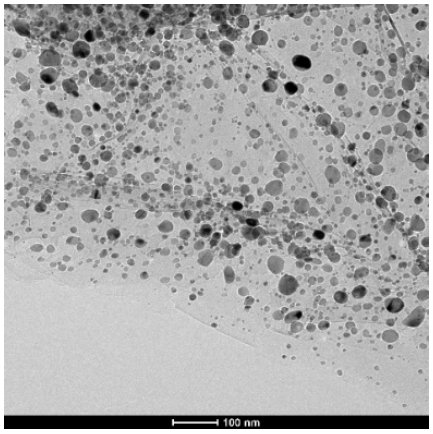

Fig. S1 TEM image of PdNi/GA catalyst.

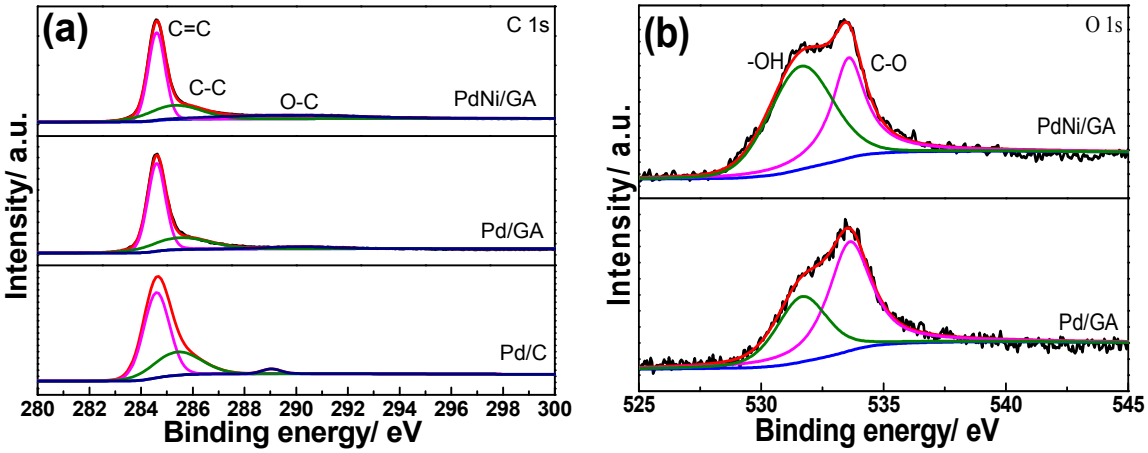

Fig. S2 (a) XPS spectra of C 1*s* of PdNi/GA, Pd/GA and Pd/C catalysts, (b) XPS spectra of O 1*s* of PdNi/GA and Pd/GA catalysts.

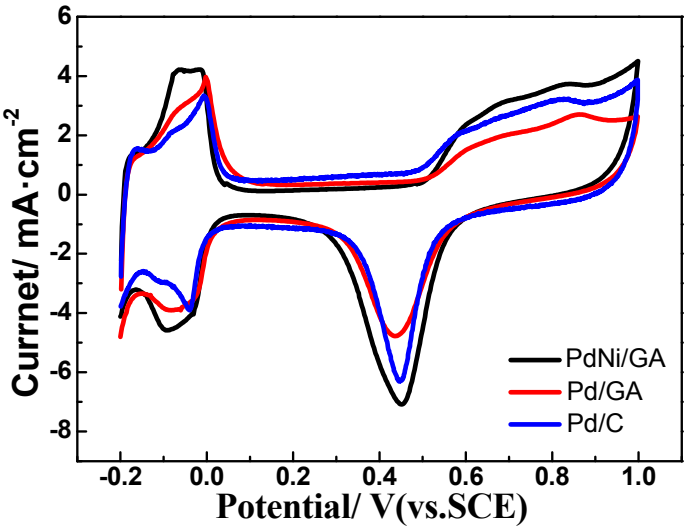

Fig. S3 Cyclic voltammograms of PdNi/GA, Pd/GA and Pd/C in 0.5 mol·L^{-1} H_2SO_4 solution at the scan rate of 50 mV·s^{-1}.

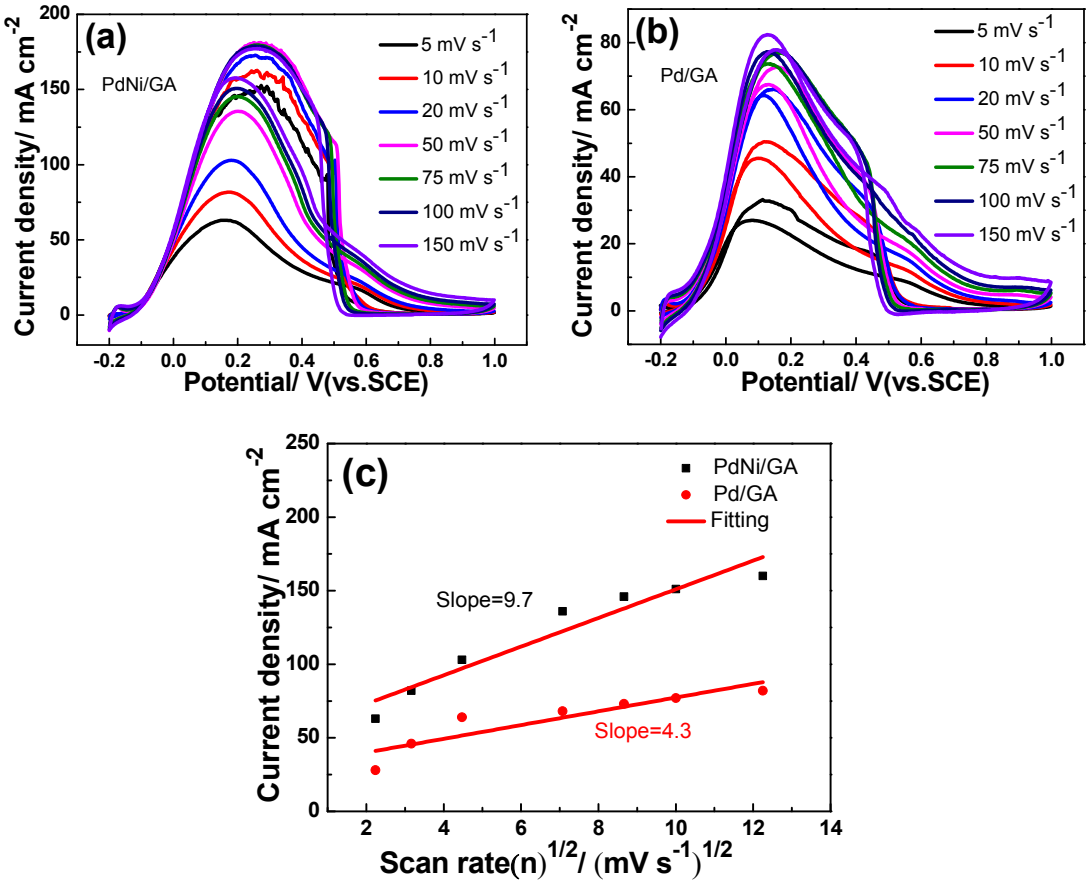

Fig. S4 Cyclic voltammograms in 0.5 mol·L^{-1} H_2SO_4/0.5 mol·L^{-1} HCOOH solution at scan rates of 5, 10, 20, 50, 75, 100 and 150 mV s^{-1} for PdNi/GA (a) and Pd/GA (b) catalysts. (c) The relationship between the peak current and the square root of scan rates of PdNi/GA and Pd/GA catalysts.

The relationship between the peak current and the square root of scan rates complies with the following equation: $i_p = 2.99 \times 10^5 n(\alpha n')^{1/2} A C_\infty D_0^{1/2} v^{1/2}$. Where i_p is the peak current, n is the electron-number for the total reaction, n' is the electron-number transferred in the rate-determining step, α is the electron transfer coefficient of the rate-determining step, A is the electrode surface area, C_∞ is the bulk concentration of the reactant, D_0 is the diffusion coefficient, v is the potential scan rate. In this paper, the slope of the i_p *vs* the square scan rate is $2.99 \times 10^5 n(\alpha n')^{1/2} C_\infty D_0^{1/2} v^{1/2}$. In the same electrolyte and the same reaction, the parameters n, C_∞ and D_0 are constant; therefore, the slope is decided by $\alpha n'$ [1]. The larger the slope value, the higher the catalytic kinetics.

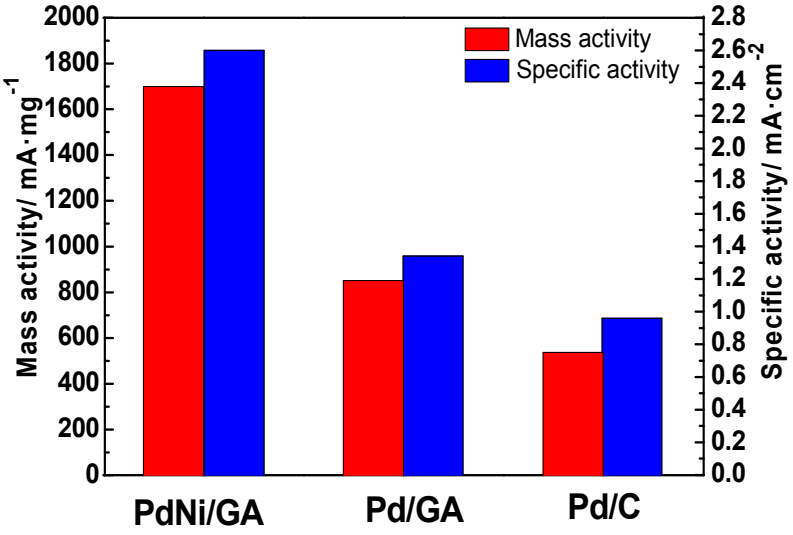

Fig. S5 Mass activity and specific activity of PdNi/GA, Pd/GA and Pd/C catalysts.

Table S1 The atomic percentage of each element on the PdNi/GA surface.

Element	Atomic percentage
C	89.86
O	8.34
Pd	0.89
Ni	0.91

Table S2 The atomic percentage of each signal of the C component.

Catalysts	Assignment	Relative intensity/ %
PdNi/GA	C－C	55
	C＝C	25
	C－O	20
Pd/GA	C－C	67
	C＝C	27
	C－O	6
Pd/C	C－C	74
	C＝C	23
	C－O	3

Table S3 The binding energy and the atomic percentage of each signal of the Pd component.

Catalysts	Assignment	Binding energy/eV	Relative intensity/%
PdNi/GA	Pd(0)	335.7	76
		340.9	
	Pd(Ⅱ)	336.9	24
		342.1	
Pd/GA	Pd(0)	335.5	61
		340.7	
	Pd(Ⅱ)	336.6	39
		341.8	
Pd/C	Pd(0)	335.3	76
		340.5	
	Pd(Ⅱ)	336.5	24
		341.7	

Table S4 Performance comparison with other reported similar catalysts.

Catalysts	Current density ($mA \cdot mg^{-1}_{Pd}$)	Reference
Pd/CMK-8-I	486	2
$Pd_{51}Cu_{49}$	517	3
Pd_1Ni_1-NNs/RGO	604.3	4
Pd/NS-G	501.8	5
PdSnAg/C	629.86	6
PdCuSn/CNTs	534.83	7
PdNiCu/C	792	8
Pd_3Fe/C	696.4	9
PdAu/C	371	10
$Pd@Ni_{1.61}B_{0.0199}$/C	400	11
Pd/C-S	1622	12
SCR-CuPdAu/C	1070	13
$PdH_{0.43}$	1060	14
Pd_4Sn NCNs	850.47	15
PdCo Nds-RGO	1362	16
Au@AuPd NCs	1250	17
Pt-Pd/PFCA/GC	1225	18
$Pd_{0.6}$-$Mn_{0.4}$	1050	19
dendritic Pd	1080	20
PdNi/GA	1699	This work

Table S5 EIS fitting parameters from equivalent circuits for different catalyst samples in 0.5 mol·L^{-1} H_2SO_4/0.5 mol·L^{-1} HCOOH.

Catalysts	R_S/Ω	CPE/S·s^{-n}	n/0 < n < 1	R_{CT}/Ω	L/H	R_0/Ω
PdNi/GA	6.8	E-3	0.7	206	4.3E-17	100
Pd/GA	6.7	3E-3	0.6	315	2.8E-17	219
Pd/C	6.8	2.8E-4	0.8	478	5.3E-4	754

Table S6 Electrochemical surface area (ECSA) estimated from CO stripping experiments, the peak potential and the onset potential for CO oxidation for the relevant catalysts.

Catalysts	ECSA/m^2·g^{-1}	Peak Potential/V *vs* SCE	Onset Potential/V *vs* SCE
PdNi/GA	65.12	0.67	0.49
Pd/GA	63.50	0.67	0.61
Pd/C	56.00	0.71	0.69

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