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Supporting information

Reconstruction of Rh nanoparticles in methanol oxidation reaction

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Fig. S1 TEM images and size distributions of Rh-ncs obtained by varying the precursor concentration: (a) 1.25mM; (b) 2.5mM; (c) 5mM; (d) 10mM; (e) 20mM; (f) 40mM, respectively.



Fig. S2 XPS spectra recorded of different types of Rh-ncs/SiO₂ catalysts. (a) fresh 2.4 nm Rh-ncs/SiO₂, (b) fresh 5.9 nm Rh-ncs/SiO₂, (c) 5.9 nm Rh-ncs/SiO₂ that was used at 453 K, (d) 5.9 nm Rh-ncs/SiO₂ that was used with temperature-programmed ramp up to 503 K, (e) 5.9 nm Rh-ncs/SiO₂ was calcined at 503 K.



Fig. S3 TEM images of Rh-ncs/SiO₂ catalysts with the average side length of 5.9 nm under various treatments. (a) before reaction; (b) after the reaction at 473 K; The average particle size of figure B is 6.5 nm.



Catalyst (5.9 nm)	Side length (a.u.)		
	normalized length ratio	"a"	"c"
fresh catalyst	0.37	68	185
used catalyst (453 K)	0.58	88	153
used catalyst (473 K)	0.9	91	101

Fig. S4 Two-dimensional diagram of (100) and (111) facets in Rh-ncs.

area (100)=6($a^2+2\sqrt{2}ac+c^2$), area (111)= $2\sqrt{3}a^2$.



Fig. S5 HR-TEM images and the interplanar spacing of 5wt% Rh/SiO₂-imp catalyst.



Fig. S6 The activity and selectivity of two Rh-ncs/SiO₂ catalysts (A, B, with the average side length of 2.4 nm, C, D, with the average side length of 5.9 nm) in the methanol oxidation reaction at 453 K for A, C, and 473 K for B, D. Oxygen content was about 25% and balanced with N_2 .



Fig. S7 ODH activity of two Rh-ncs/SiO₂ catalysts. The reaction condition was as the same as upper figure. (The irreversible nature of the oxidative conversion of CH_3OH to HCHO requires one oxidative CH_3OH dehydrogenation (ODH) event for each HCHO, DMM, and MF molecule formed. ODH rates rigorously reflect the intrinsic oxidation reactivity of active domains, without contributions from methanol molecules consumed because of various secondary reactions.)



Fig. S8 The interplanar spacing of Rh ncs in HR-TEM images (Figure 6).



Fig. S9 Stability of impregnation-derived Rh/SiO₂ catalyst.