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Atomic Dispersed Pd site on Ti-SBA-15 for Efficient Catalytic Combustion of Typical Gaseous VOCs

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Synthesis of Pd0.5 NPs/Ti-SBA-15 catalyst

Reference sample, Pd0.5 NPs/Ti-SBA-15, was synthesized by using $NaBH_4$ as reducing agent. Typically, a certain amount of Pd precursor was added into the PVP (Pd/PVP mass ratio=1:1.2) in the deionized water, stirring for 6 h. Then a moderate amount of Ti-SBA-15 dispersed into methanol was added into the mixed solution. The mixture was further vigorously stirred for 3 h. Then, a considerable amount of $NaBH_4$ dissolved in deionized water was dropwise added. After continuously stirring for 1 h, the solid sample was filtered and dried at 60 °C in an oven for overnight. Finally, the solid sample was calcined at a ramp of 2 °C/min from room temperature to 500 °C and kept at this temperature for 3h in a muffle furnace, hence obtaining the Pd0.5 NPs/Ti-SBA-15 catalyst. The actual loading amount of Pd0.5 NPs/Ti-SBA-15 was determined by ICP-AES to be 0.52 wt%.

DFT calculation

The adsorption energy (E_{ads}) of styrene, cyclohexane and n-hexane on the Pd site was calculated basing on the adsorption configuration a:

$$E_{ads} = E_{VOC/surface} - (E_{VOC} + E_{surface})$$

where $E_{surface}$ is the total energy of adsorption configuration a, and the $E_{VOC/surface}$ is the total energy of the VOC/surface. While, the E_{VOC} is the free energy of styrene, cyclohexane and n-hexane. The more negative value from this equation indicated the stronger adsorption accordingly.

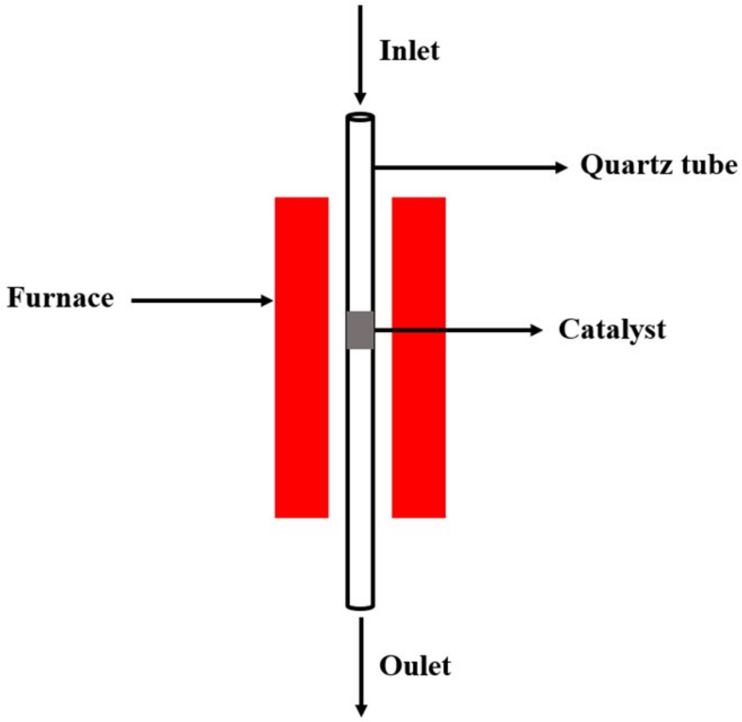

Fig. S1. Schematic diagram of the thermal reactor.

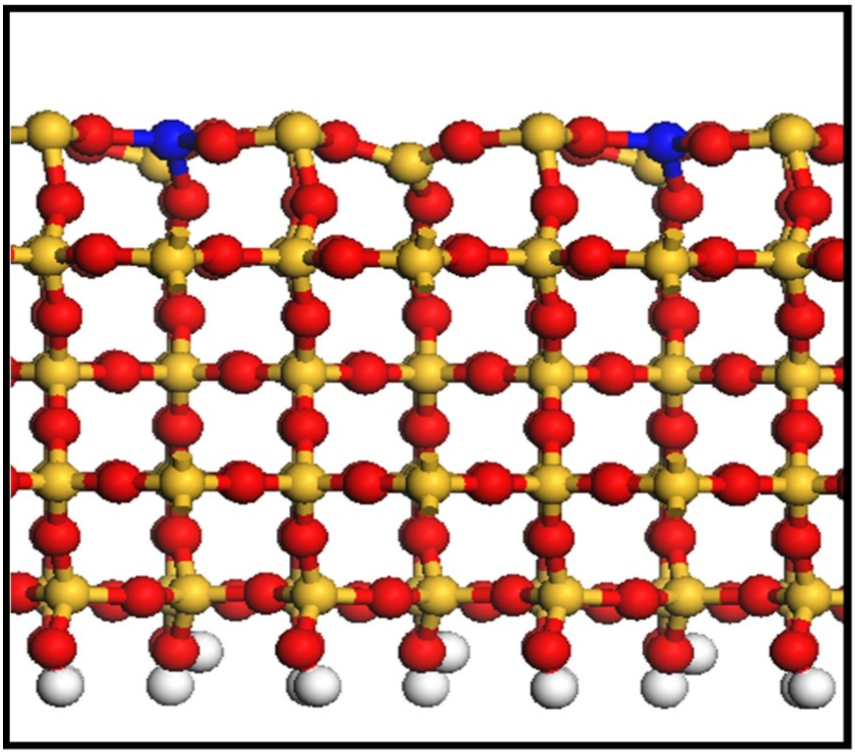

Fig. S2. Optimized configurations of the Ti-SBA-15. The red, yellow, blue and white spheres represent O, Si, Ti and H atoms, respectively.

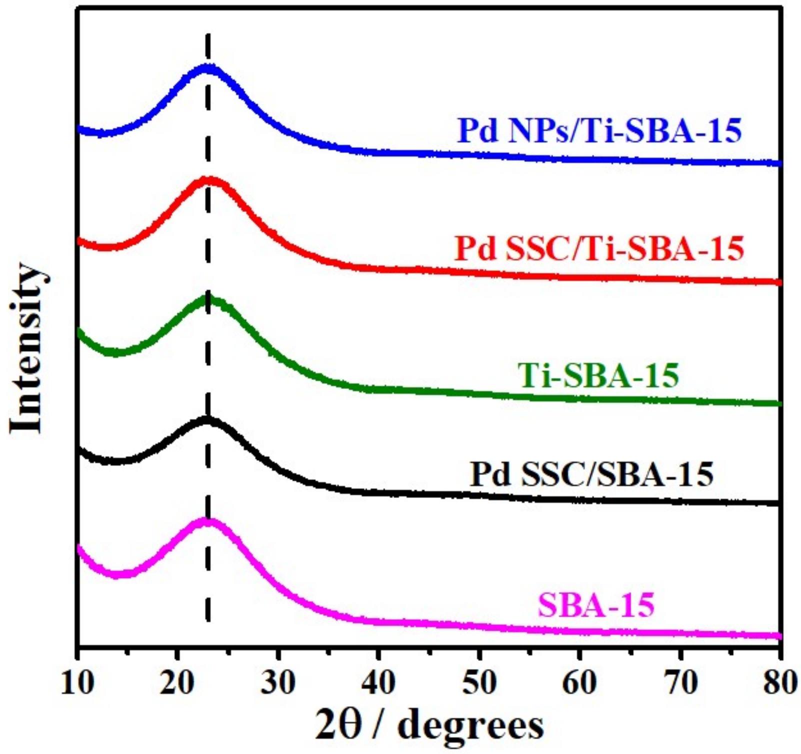

Fig. S3. Wide-angle XRD patterns of the as-prepared samples.

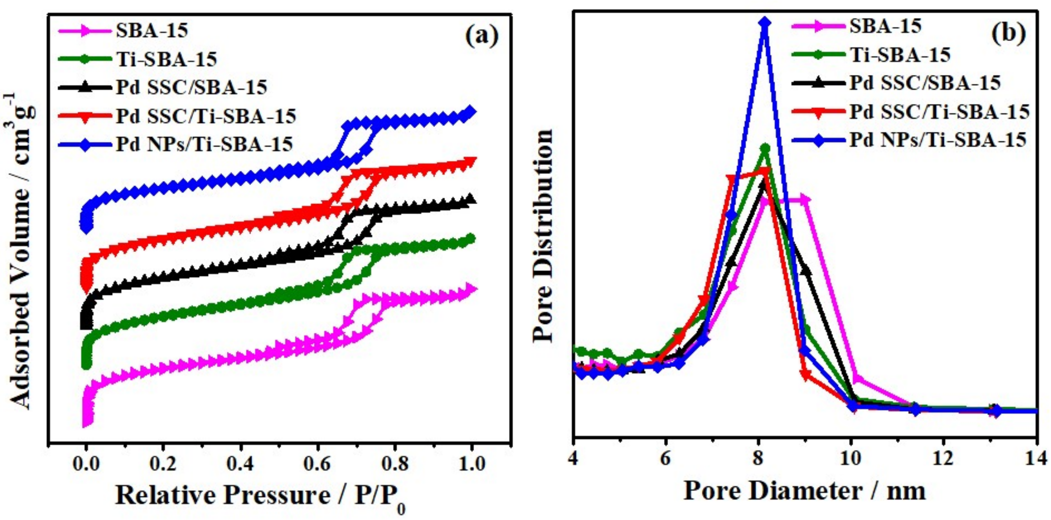

Fig. S4. (a) N_2 adsorption-desorption isotherms and (b) Pore size distributions of as-prepared samples.

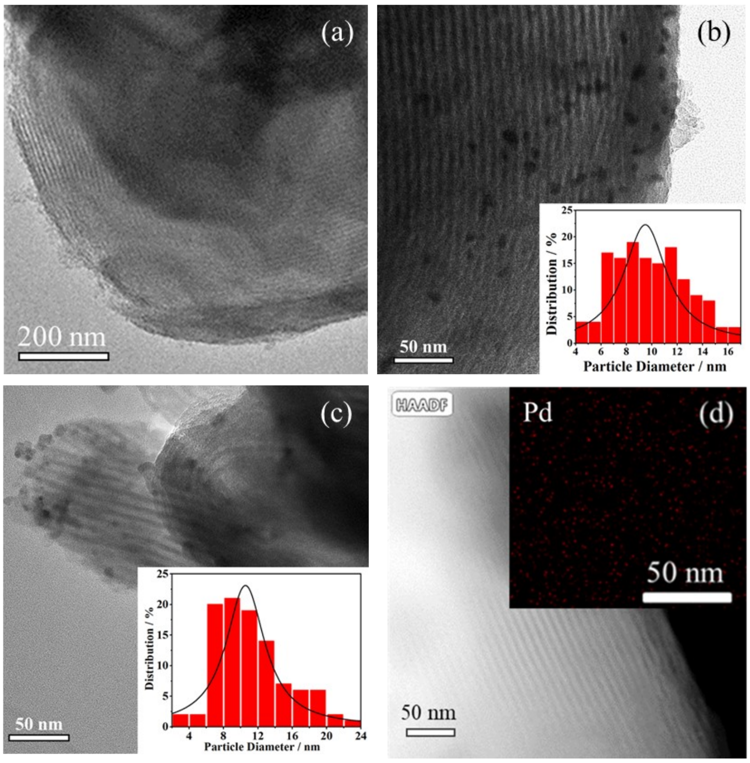

Fig. S5. HR-TEM images of (a) Pd SSC/SBA-15, (b) Pd NPs/Ti-SBA-15, (c) Pd0.5 NPs/Ti-SBA-15 and the size distribution images of Pd nanoparticles of Pd NPs/Ti-SBA-15 and Pd0.5 NPs/Ti-SBA-15 sample (inset) and (d) HADDF-STEM and EDX elemental mapping of Pd (inset) of recycled Pd SSC/Ti-SBA-15.

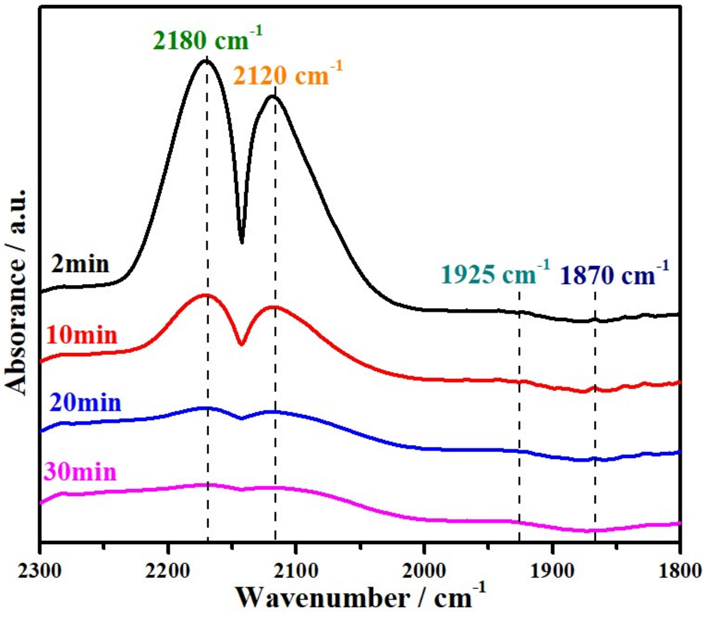

Fig. S6. In-situ DRIFT spectra of CO adsorbed on Pd NPs/Ti-SBA-15 with different time in the N_2 flow (30.0 mL/min) at 30 °C.

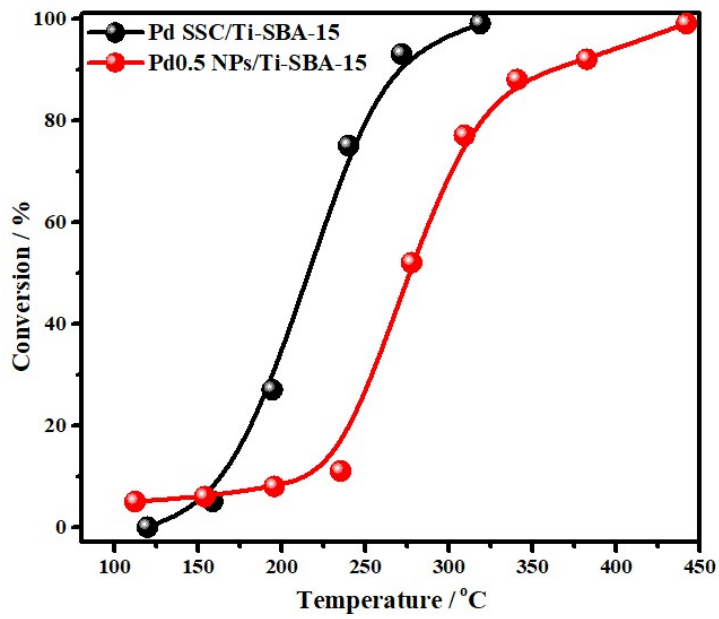

Fig. S7. Cyclohexane conversion as a function of temperature over Pd SSC/Ti-SBA-15 and Pd0.5 NPs/Ti-SBA-15.

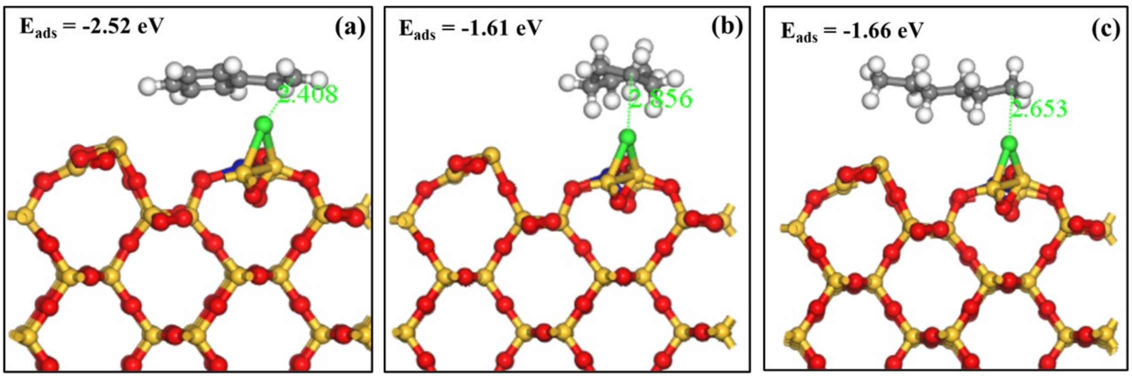

Fig. S8. Configurations for styrene (a), cyclohexane (b) and n-hexane (c) adsorption on the Pd site of Pd SSC/Ti-SBA-15. The red, yellow, blue, green and white spheres represent O, Si, Ti, Pd and H atoms, respectively.

Table S1. Textural parameters of various prepared samples.

Sample	Surface area	Pore diameter	Pore volume	Pd content (wt%)	Ti content (wt%)
SBA-15	1024.081	8.984	0.760	0	0
Ti-SBA-15	911.034	8.318	0.766	0	0.14
Pd SSC/SBA-15	891.906	8.117	0.708	0.05	0
Pd SSC/Ti-SBA-	859.942	8.315	0.683	0.04	0.14
Pd NPs/Ti-SBA-	770.437	8.124	0.753	0.05	0.13

Table S2. Different conversions temperatures of cyclohexane onto the prepared samples.

Catalyst	T_{50} (°C)	T_{90} (°C)
Pd SSC/SBA-15	340	420
Pd SSC/Ti-SBA-15	220	270
Pd NPs/Ti-SBA-15	350	500
Pd0.5 NPs/Ti-SBA-15	275	370

Table S3. Adsorption energies of Pd atom on different configurations over Pd SSC/Ti-SBA-15.

Configurations	Eads (eV)
a	-4.95
b	-4.57
c	-4.52

Table S4. Different conversions temperatures of different VOCs on Pd SSC/Ti-SBA-15.

VOC	T_{50} (°C)	T_{90} (°C)
Styrene	180	210
Cyclohexane	220	270
n-hexane	275	310

Table S5. Recent works of catalytic VOCs combustion over different catalysts.

Sample	VOC	Concentration ppm	Loading Amount (wt%)	T_{50} (°C)	T_{90} (°C)	Ref
Pt/Al_2O_3	cyclopentane	6200	0.26	312	-	1
Au/TiO_2	n-hexane	125	3	350	-	2
Pt/Al_2O_3	n-hexane	1500	0.12	300	400	3
$Ce_{0.99}Cu_{0.01}O_2$	n-hexane	260	-	-	400	4
CuO/Al_2O_3	styrene	1000	8.2	240	325	5
$PtCu/CeO_2$	n-pentane	50	0.08	300	400	6
Pd/SBA-15	toluene	650	0.3	261	292	7
Pd/C	toluene	1000	0.5	280	360	8
Pd/TiO_2	toluene	1000	0.5	230	250	9
Pd/ V_2O_5/Al_2O_3	benzene	482	5	280	310	10
Pd SSC/Ti-SBA-15	styrene	50	0.04	180	210	this work
Pd SSC/Ti-SBA-15	cyclohexane	40	0.04	220	270	this work
Pd SSC/Ti-SBA-15	n-hexane	40	0.04	275	310	this work

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