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Temperature-induced structure reconstruction to prepare thermally stable single-atom Pt catalyst

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Abstract: Single-atom noble metal on catalyst support tend to migrate and agglomerate into nanoparticles due to high surface free energy at elevated temperatures. Temperature-induced structure reconstruction of support can firmly anchor single-atom Pt species to adapt high-temperature environment. We used Mn₃O₄ as a restructurable support to load single-atom Pt and further turned into single-atom Pt-on-Mn₂O₃ catalyst via high-temperature treatment, which is extremely stable under calcination conditions of 800°C for 5 days in humid air. Experiments reveal that high valence Pt⁴⁺ with more covalent bonds on Mn₂O₃ are essential for anchoring isolated Pt atoms by strong interaction. Optimized catalyst by moderate H₂O₂ etching exhibits the best catalytic performance and the excellent thermal stability of single-atom Pt-Mn₂O₃ interaction.

Oxide-supported noble-metal catalysts have received tremendous attention due to their extensive utilization in many fields such as chemical synthesis, energy conversion and environmental purification. Although the introduction of noble metal plays a critical role in enhancing reactivity and changing selectivity, the relatively low natural abundance and expensive price of noble metal hinder the large-scale application of such catalysts. The appearance and development of single-atom catalysts (SACs) provides a better alternative strategy because of its maximized noble metal atom-utilization efficiency.^[11] Unique metal coordination environment makes SACs to show more remarkable catalytic behavior in specific reaction than normal noble-metal catalysts.^[22] To obtain SACs, several innovative synthetic strategies have been developed including thermal emitting, ball milling, atomic layer deposition, and wet-chemistry approaches.^[3] Nevertheless, single-atom noble metals tend to aggregate into nanoclusters (NCs) or nanoparticles (NPs) at high-temperature reactions, resulting in the noteworthy reduction of catalytic activity.^[4] Hence, stabilizing noble metals on catalyst support in single-atom status remains a formidable challenge.

At high-temperature, the difference in free energy and density of adatoms drives the migration of single atoms to NPs, which is the main reason for the sintering of SACs and called Ostwald Ripening.^[5] To resist sintering of SACs, noble metal atoms trapping on defective metal oxides (such as CeO₂) was proposed, where atoms loaded on the defects have relatively low free energy and maintain the stability of noble metal single atom at high-temperature.^[1b, 6] However, the density of defects and surface area of support have great effect on concentration of single atoms, so that the preparation of thermally-stable SACs with high-density noble metal single atoms is more difficult. Recently, Wei et al. and Lang et al. reported high-density and thermally stable SACs synthesized by the reversed sintering process, in which noble metal particles can

convert into single atoms at high-temperature based on the thermal behavior of supported noble metal particles.^[7] Although it is achieved to obtain SACs, the conversion process of particles to atoms at high-temperature is commonly uncontrollable. Therefore, we think about the possibility to determine thermally-stable SACs through using controllable thermal behavior of support. Additionally, it is more necessary to validate the stability of single-atom dispersion of SACs for a long time at high temperature in humid air or reaction atmosphere.

Herein, we report a new synthetic strategy of superior thermally-stable platinum SACs through structure reconstruction of MnO_x support at elevated temperature. We utilized Mn₃O₄ as a preloading support for single-atom Pt that could firmly anchor Pt atoms on support in the following phase transition to Mn₂O₃ induced by hightemperature treatment. We observed that confined Pt atoms on Mn₂O₃ converted into high-valent Pt⁴⁺ species with strong interaction,^[7b, 8] and still existed in single-atom dispersion status even through calcination at 800 °C with 3 vol.% water vapor for 5 days. However, the large Pt NPs would be formed under the same conditions only for 5 hours on unreconstructed Mn₂O₃ with single-atom Pt. These results can be confirmed by x-ray diffraction (XRD), energy dispersive spectroscopy (EDS), aberrationcorrected scanning transmission electron microscopy (AC-STEM), diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS), X-ray absorption near edge structure (XANES) and X-ray absorption fine structure (EXAFS). Additionally, optimization through moderate H₂O₂ etching, the temperature-induced structure reconstructed Pt-on-Mn₂O₃ SACs also showed the outstanding thermal stability and stable catalytic activity in high-temperature methane oxidation.

Our previously reported redox precipitation method was applied to preload singleatom Pt species on the as-obtained Mn_3O_4 and Mn_2O_3 , denoted as Pt_1/Mn_3O_4 and

Pt1/Mn2O3, respectively.^[9] Isolated Pt single atoms on both Mn3O4 and Mn2O3 are initially detected by AC-STEM, where Pt species appear as highly-dispersed distribution with high density on the surface of Mn₃O₄ and Mn₂O₃ and no Pt NCs or NPs are observed in Pt₁/Mn₃O₄ and Pt₁/Mn₂O₃ (Figure 1A and 1C). CO-DRIFTS spectra display the coverage-independent band at around 2099 cm⁻¹ ascribed to CO linearly bound to isolated Pt atoms on both Pt₁/Mn₃O₄ and Pt₁/Mn₂O₃ (Figure S1), which indirectly proves the prevalence existence of single-atom Pt on catalyst support.^[10] Subsequent thermal treatment over both Pt₁/Mn₃O₄ and Pt₁/Mn₂O₃ was carried out at high-temperature, as illustrated in Figure 1. Pt₁/R-Mn₂O₃-5D was obtained by calcining Pt₁/Mn₃O₄ at 800°C in air with 3 vol.% water vapor for 5 days and its AC-STEM image obviously shows the single-atom dispersion of Pt (Figure 1B), which indicates the ultra-stability of Pt single atoms in the calcination process. Element distribution images of Pt₁/R-Mn₂O₃-5D display the uniform distribution of Pt element in EDS mapping (Figure S2), which implies the highly-dispersed state of Pt species on the support. XRD pattern collected for Pt₁/R-Mn₂O₃-5D (Figure 2A) displays only the characteristic peaks of Mn₂O₃ phase from high-temperature-induced reconstruction of Mn₃O₄, which is in accordance with the phase change of pure support (Figure S3). No Pt-phase characteristic peaks appear in XRD pattern, which also verifies only highlydispersed Pt species existed on Mn₂O₃. The interaction between single-atom Pt and support in Pt₁/Mn₃O₄ is extremely enhanced due to the generation of enough surface defects and oxygen vacancies in conversion of Mn_3O_4 to Mn_2O_3 , which can firmly trap and confine single-atom Pt and result in ultra-thermal stable Pt single atoms on the surface of Mn_2O_3 in $Pt_1/R-Mn_2O_3-5D$. Conversely, non-reconstruction Mn_2O_3 as the support cannot stabilize Pt species at single-atom dispersion. Only 5 hours treatment on Pt₁/Mn₂O₃ at 800°C in air with 3 vol.% water vapors to obtain Pt_{NP}/Mn₂O₃-5H, it leads

to single-atom Pt to migrate and agglomerate into large-size nanoparticle of about 13.8 nm (Figure 1D). It is also confirmed by XRD pattern of Pt_{NP}/Mn_2O_3 -5H (Figure 2A), in which a small peak assigned to lattice plane of Pt (111) appears at 2θ = 39.7° (JCPDF no. 01-087-0646) and the grain size is calculated to be 11.8 nm by Scherrer equation.^[1b] EDS images of Pt_{NP}/Mn₂O₃-5H (Figure S4) reveal the presence of Pt NPs on the surface. Thus, we can deduce that the reconstruction process of Mn₃O₄ to Mn₂O₃ firmly anchors Pt species in single-atom status. Furthermore, this synthetic method also successfully fulfils ultra-stable single-atom Pt species with high density on the support with very low surface area, as obtained Mn₂O₃ and Mn₃O₄.



Figure 1. A) AC-STEM image of Pt₁/Mn₃O₄. B) AC-STEM image of Pt₁/R-Mn₂O₃-5D obtained *via* calcining Pt₁/Mn₃O₄ at 800°C in air with 3 vol.% water vapor for 5 days. C) AC-STEM image of

 Pt_1/Mn_2O_3 . D) HRTEM image of Pt_{NP}/Mn_2O_3 -5H obtained *via* calcining Pt_1/Mn_2O_3 at 800°C in air with 3 vol.% water vapor for 5 hours.

XANES measurements (Figure 2B) confirm that the chemical state of Pt species in Pt₁/R-Mn₂O₃-5D strongly resembles PtO₂, implying a high valence state of Pt⁴⁺ surrounded by oxygen and a larger population of unoccupied Pt 5d states for Pt ions in Pt₁/R-Mn₂O₃-5D.^[7b, 11] Fourier transform radial distribution functions of k3-weight EXAFS spectrum for Pt₁/R-Mn₂O₃-5D (Figure 2C) shows a peak at around 1.60 Å from the Pt-O contribution and no peak is observed for Pt-Pt bond contribution. Additionally, a second shell peak at around 2.81 Å that is distanced from *R* values of Pt foil (2.38 Å) and PtO₂ (2.67 Å) is attributed to the Pt–Mn coordination in Pt₁/R-Mn₂O₃-5D. As a result, we deduced that Pt-O and Pt–Mn coordination are from the strong interaction between the Pt atom and Mn₂O₃ due to the firm anchoring of high-valent single-atom Pt on the surface or sub-surface of support *via* structure reconstruction, which effectively inhibits migrating and sintering of Pt atom.^[12]

catalyst	O _{latt} /O _{ads}	Pt species (Pt-4f) (%) ^[a]			Mn species (Mn-2p) (%) ^[a]		
		Pt^{4+}	Pt^{2+}	Pt^0	Mn^{4+}	Mn ³⁺	Mn^{2+}
Pt ₁ /Mn ₃ O ₄	0.70	81.8	18.2	0.0	16.1	22.2	61.7
Pt_1/Mn_2O_3	1.52	55.1	44.9	0.0	16.7	50.6	32.7
$Pt_1/R-Mn_2O_3-5D$	2.11	100.0	0.0	0.0	17.1	60.2	22.8
Pt _{NP} /Mn ₂ O ₃ -5H	2.09	54.7	17.1	28.3	22.3	51.2	26.5

Table 1: Summary of XPS Anal	lysis
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[a] The relative contents of Pt, O and Mn species are calculated according to areas of simulated peaks in Pt-4f7/2, O-1s and Mn-2p5/2 regions, respectively.

XPS patterns (Figure 2D) of Pt-4f for Pt_1/Mn_3O_4 and Pt_1/Mn_2O_3 show the four well-fitted peaks with binding energies of 77.9, 75.8, 74.5 and 72.4 eV, which are attributed to $Pt^{4+}4f_{5/2}$, $Pt^{2+}4f_{5/2}$, $Pt^{4+}4f_{7/2}$, and $Pt^{2+}4f_{7/2}$, respectively. After high-

temperature calcination, the derived Pt₁/R-Mn₂O₃-5D only remains two peaks at around 78.2 and 74.8 eV from the spin-orbital split of Pt⁴⁺, which indicates that hightemperature treatment leads to complete conversion of Pt²⁺ to Pt⁴⁺ (Table 1).^[13] For Pt_{NP}/Mn₂O₃-5H, more complicate chemical valence of Pt in XPS Pt-4f curve are observed that displays the inherited Pt⁴⁺ and Pt²⁺ peaks and a new metallic Pt⁰ species with the binding energies of 74.3 and 71.1 eV. Synchronously, an obvious increasing ratio of Mn³⁺ in Mn-2p XPS of Pt₁/R-Mn₂O₃-5D (Figure S5 and Table 1) explains the occurrence of structure reconstruction of support from Mn₃O₄ to Mn₂O₃. However, the XPS ratio of Mn³⁺ in Pt_{NP}/Mn₂O₃-5H keeps the same level with Pt₁/Mn₂O₃, which suggests Mn₂O₃ support is more stable under calcination. Structure reconstruction of support causes that surface dispersed Pt single atoms of Pt₁/Mn₃O₄ are deeply confined on the surface or subsurface of Mn₂O₃ in Pt₁/R-Mn₂O₃-5D during calcination. It facilitates the generation of more Pt-O bonds between single-atom Pt and O of Mn₂O₃ and convert all Pt²⁺ to Pt⁴⁺, which further enhances the interaction between Pt and Mn₂O₃ to stabilize the single-atom structure at least for 5 days. Due to lack of confinement effect, a large amount of Pt⁴⁺ and Pt²⁺ existed on the surface of Pt₁/Mn₂O₃ at elevated temperature easily generates the volatile gaseous PtO₂ that can depart from the surface of support and trap each other to form larger NPs (Ostwald Ripening).^[7b] It is also confirmed by HRTEM and XRD analysis of Pt_{NP}/Mn₂O₃-5H, which is quite consistent with the sintering of Pt to NPs on support after high-temperature calcination.^[6, 14] Based on analysis of XPS data, Pt/Mn ratios on the surface of Pt₁/Mn₃O₄ and Pt₁/R-Mn₂O₃-5D are calculated, which display an obvious decreasing of surface Pt content from 2.7:97.3 of Pt₁/Mn₃O₄ to 0.7:99.3 of Pt₁/R-Mn₂O₃-5D via calcination (Table S1). However, inductively coupled plasma-optical emission spectroscopy (ICP-OES) results of Pt contents in Pt₁/Mn₃O₄ and Pt₁/R-Mn₂O₃-5D

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express that they have the same Pt loading of 0.82 wt% in the bulk (Table S1). This discrepancy comes from that structure reconstruction of support makes some Pt single atoms locate in sub-surface region of support and is beyond the scope of XPS detection. The very close Pt contents of ICP-OES result for Pt₁/Mn₂O₃ and Pt_{NP}/Mn₂O₃-5H prove that Pt has not been noticeably lost in vaporization depletion *via* the path of volatile PtO₂ at high-temperature.



Figure 2. A) XRD patterns of samples in full range. Except Pt_{NP}/Mn_2O_3 -5H, no peaks corresponding to Pt species are observed for the other samples. The inset showing an obvious peak at 39.7° from Pt (111) in the curve of Pt_{NP}/Mn_2O_3 -5H confirms the existence of Pt NPs. B) Pt L_{III} XANES spectrum of Pt_1/R -Mn₂O₃-5D alongside PtO₂ and Pt foil references. C) Fourier transform radial distribution function of the Pt L_{III}-edge k3-weighted EXAFS spectra of Pt_1/R -Mn₂O₃-5D in

comparison with PtO_2 and Pt foil. D) Pt-4f XPS spectra of Pt_1/Mn_3O_4 , $Pt_1/R-Mn_2O_3-5D$, Pt_1/Mn_2O_3 and Pt_{NP}/Mn_2O_3-5H .

The cationic oxidation state on transition metal oxides can be changed during structure reconstruction especially for Mn cation with multiple oxidation states.^[15] Temperature-induced structure reconstruction in air inserts more oxygen species into crystal phase structure of Mn₃O₄ during the irreversible phase transition of Mn₃O₄ to Mn₂O₃ due to charge compensation in the conversion of low-valence Mn ions to high-valence Mn ions. In this process, the originally ordered structure of Mn₃O₄ is destroyed, which inevitably forms surface deformation and generates structure defects to firmly confine the already-existed Pt single atoms. It finally results in the formation of the strong interaction between Pt atoms and support by more coordination bonds of Pt⁴⁺ and oxygen as well as surface-layer confinement.

Generally, noble metal species of supported noble metal catalyst as catalytic sites plays a key role in catalytic reaction. Thus, to expose more single-atom Pt catalytic sites in Pt₁/R-Mn₂O₃-5D, chemical etching by H₂O₂ to react with MnO_x to form watersoluble Mn²⁺ was used to prepare xH_2O_2 -Pt₁/R-Mn₂O₃-5D (x = 0.25, 0.5, 1.0 mL) based on our previous research (Figure 3A).^[9a] No change can be obviously observed in XPS results after treatment by different amount of H₂O₂ except the gradually enhanced intensity of Pt⁴⁺ signals (Figure S6 and Table S2), which reflects surface Pt single atoms are increasingly exposed accompanying with the removal of covered MnO_x. CO-DRIFTS of xH_2O_2 -Pt₁/R-Mn₂O₃-5D show the coverage-independent band at around 2080-2095 cm⁻¹ (Figure 3B), which is the only feature attributed to CO linearly bound to isolated Pt atoms.^[1b, 16] The raised intensity of the peak with increasing H₂O₂

amount explains that more Pt single atoms appear on the surface of Mn_2O_3 after H_2O_2 treatment and linearly coordinate with more CO. ICP-OES results confirm that samples still keep very close Pt contents although through H_2O_2 etching (Table S1), indicating single-atom Pt on Mn_2O_3 are not easily washed off.



Figure 3. A) H_2O_2 etching to remove Mn oxides covered on Pt single atoms. B) DRIFTS spectra of $Pt_1/R-Mn_2O_3-5D$ and $xH_2O_2-Pt_1/R-Mn_2O_3-5D$. C), D) CH₄ conversion (C) and CO₂ yield (D) on $Pt_1/R-Mn_2O_3-5D$, $xH_2O_2-Pt_1/R-Mn_2O_3-5D$ and Mn_2O_3 .

We test the catalytic activity of methane oxidation on catalyst to verify whether the catalyst etched by H_2O_2 show higher catalytic reactivity. High-temperature catalytic

behavior of CH₄ oxidation on xH_2O_2 -Pt₁/R-Mn₂O₃-5D (Figure 3C and 3D) presents that 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D owns the best catalytic activity for CH₄ oxidation with 50% of conversion at 518°C (T_{50%}) and 90% of conversion at 603°C (T_{90%}), which are more super than untreated Pt₁/R-Mn₂O₃-5D (T_{50%} = 546°C and T_{90%} = 639°C) and much better than pure Mn₂O₃ (T_{50%} = 580°C and T_{90%} = 694°C) (Table S3). Noticeably, more H₂O₂ addition in preparation of xH_2O_2 -Pt₁/R-Mn₂O₃-5D is helpless to further promote catalytic activity. As observed for 1.0H₂O₂-Pt₁/R-Mn₂O₃-5D, it shows the slightly less catalytic activity than that of 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D. It manifests moderate exposed Pt single atoms as well as synchronously owning the optimized strong Pt-support interaction on the surface of Mn₂O₃ can facilitate CH₄ catalytic oxidation. Actually, it is limited to distinguish the characteristic difference in these catalysts. As a comparison with Pt₁/R-Mn₂O₃-5D, 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D is almost unchanged in properties of XRD, AC-STEM, XANES and EXAFS (Figure 4B, 4D, 4G and Figure S7) except the intensity of XPS Pt⁴⁺ peaks (Figure 4E).

We also performed the cycle experiment of CH_4 oxidation on $0.5H_2O_2$ -Pt₁/R-Mn₂O₃-5D at alternative reaction temperatures for 3 cycles (700 and 500°C, each temperature stage for 10 h, Figure 4A), exhibiting the stable catalytic activity in both high and low conversions for at least 60 h. The structure stability of 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D in oxidation reaction is investigated by analysis of AC-STEM, XANES, EXAFS, XPS, XRD and ICP-OES before and after the cycle experiment. After 60-hour reaction, the used catalyst 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used still retains the single-atom dispersed Pt on Mn₂O₃ imaged in AC-STEM (Figure 4B and 4C), the same coordination sphere and valence state of Pt with $0.5H_2O_2$ -Pt₁/R-Mn₂O₃-5D (Figure 4D-4G), and the almost same XRD pattern and Pt content of ICP-OES with 0.5H2O2-Pt1/R-Mn2O3-5D confirm (Figure **S**7 and Table S1). These results that the catalyst 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D has an outstanding structure stability even through the harsh high-temperature reaction. Compared with the reported Pt catalysts (Table S4), 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D with the lowest Pt loading has relatively good catalytic activity for methane oxidation and long-term catalytic stability for both low and high conversions, which is ascribed to single atom distribution of Pt and strong confinement effect of Pt atoms on the surface of support.



Figure 4. A) 60-hour high-temperature reaction stability test on 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D at 700 and 500 °C with different conversions. B), C) AC-STEM images of 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D (B) and 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used (C). D) Pt L_{III} XANES spectra of 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D and 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D-Used. E), F) XPS spectra collected at the Pt-4f on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-5D on the 0.5H₂O₃-SD on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-SD on the 0.5H₂O₂-Pt₁/R-Mn₂O₃-SD on the 0.5H₂O₃-SD on the 0.5H₂O₃-SD on the 0.5H₂O₃-SD on

 Mn_2O_3-5D (E) and $0.5H_2O_2-Pt_1/R-Mn_2O_3-5D$ -Used (F). G) Fourier transform radial distribution function of the Pt L_{III}-edge k3-weighted EXAFS spectra of $0.5H_2O_2-Pt_1/R-Mn_2O_3-5D$ and $0.5H_2O_2-Pt_1/R-Mn_2O_3-5D$ -Used.

In summary, structure reconstruction to anchor single-atom noble metals is an efficient approach to obtain high-thermal stable SACs. As a common phenomenon, the phase transition of materials is structure reengineering process, which can achieve by various means. In our research, temperature-induced phase transition process from Mn_3O_4 to Mn_2O_3 is a critical prerequisite to firmly confine the existed surface singleatom Pt species on support, which dramatically promotes the Pt-support interaction and ensures Pt species with a stable single-atom dispersion status in catalyst. The assynthesized Pt₁/R-Mn₂O₃-5D maintains the single-atom stability after calcination at 800 °C in air with water vapor for at least 5 days. Not only that, it also exhibits the outstanding structure stability *via* harsh high-temperature catalytic behavior of CH₄ oxidation on xH_2O_2 -Pt₁/R-Mn₂O₃-5D optimized by moderate H₂O₂ etching. Thus, support reconstruction provides a new direction to successfully solves the problem of the instability for noble-metal SACs at high-temperature.

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Conflict of interest

The authors declare no competing interests.

Keywords: single-atom catalyst • Pt • thermal stability • structure reconstruction • strong interaction

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Accompanying with phase transition of support, surface reconstruction process solidly confines Pt single atoms to form enhanced strong interaction of Pt and support, which restrains noble metal atoms to aggregate and evaporate at high-temperature. The resulting single-atom noble metal catalyst exhibits excellent thermal stability both at 800°C in the humid air for 5 days or in high-temperature CH₄ catalytic combustion.