

Preparation of Palladium Supported on Ferric Oxide Nano-catalysts for Carbon Monoxide Oxidation in Low Temperature

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Abstract: Catalytic property of Pd/Fe₂O₃ catalysts on carbon monoxide (CO) oxidation at low temperature were investigated in this paper. Both the as-prepared and H₂-pretreated Pd/Fe₂O₃ catalysts show catalytic performances on CO oxidation. The CO was completely converted at 333 K for the as-prepared sample, whereas at 313 K for H₂-pretreated Pd/Fe₂O₃-573 catalyst. The catalytic performance of the Pd/Fe₂O₃ catalyst decreases with increased calcination temperature. This may be due to the increased crystallinity of the support and decreased metal-support interaction. Progressive deactivation of the catalysts during long-time reaction was associated with the formation of carbonates on the catalyst surface that inhibits CO activation or intermediate transformation.

Keywords: Pd/Fe₂O₃; Carbon monoxide; Catalytic oxidation; Low temperature

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Introduction

Carbon monoxide (CO) catalytic oxidation is considered as a very important reaction in environmental applications, for example, vehicle exhausts elimination in semi-closed spaces [1-3]. To date, this reaction was mostly studied on supported noble metal catalysts like Au, Pt and Pd due to their high activities at low temperatures [4-6]. However, high prices and deactivation drawbacks of these metal catalysts limit applications in many fields [5,7-9]. The deactivation was mainly originated from aggregation of active metal, coverage of carbonates and competitive adsorption between CO and moisture [10,11]. In contrast, much lower cost Pd catalyst showed comparable activity and long lifespan for CO oxidation at low temperatures [12-14]. It has a very potential possibility that Pd catalysts might replace Au catalysts for CO oxidation, especially when the catalysts needed to be prepared in large-scale and

applied in engineering aspects.

Catalytic performance is found highly dependent on the catalyst preparation methods [15]. Co-precipitation is one of the simplest methods for preparation of homogeneous dispersed catalysts. Some results even showed single-atom dispersion that owning high intrinsic activity [16]. Pd nanoparticles can be synthesized by co-precipitation method with high dispersion, and exhibited excellent activity in CO oxidation [17]. Because of the unique redox behavior and coordinative unsaturated Fe sites, Fe₂O₃ was considered to be an effective support for catalyst preparation [18,19]. It was reported that noble metals/FeO_x are promising catalysts for CO oxidation [20].

In this paper, Pd/Fe₂O₃ catalysts were prepared by co-precipitation method and their application in CO oxidation at low temperatures was investigated. The advantages of H₂-pretreated catalysts were analyzed, the reaction mechanism was discussed and the reason

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for deactivation was illustrated.

Experimental

PdCl₂ (AR) was available from Heraeus Materials Technology Shanghai Ltd., Fe(NO₃)₃·9H₂O (AR) and Na₂CO₃ (AR) from Sinopharm Chemical Reagent Co., Ltd. (SCRC).

The 2.0 wt% Pd/Fe₂O₃ catalysts were prepared by co-precipitation method. Typically, PdCl₂ and Fe(NO₃)₃ were firstly dissolved in 60 mL de-ionic water and stirred for 30 min. Then, the mixture was slowly dropped into Na₂CO₃ aqueous solution at 338 K under vigorous stirring. The pH of the solution was maintained 8~9. After aged for 2 h, the brown precipitate was filtrated and washed by hot water for several times until the chloride ions were undetected by silver nitrate. The finally obtained slurry was dried at 373 K overnight and calcined for 4 h at 573, 673 and 773 K, respectively. The samples were noted as Pd/Fe₂O₃-*T* (*T* is the calcination temperature).

The Brunauer-Emmett-Teller (BET) specific surface area of the sample was measured by adsorption-desorption of nitrogen at 77 K using an ASAP 2020 instrument (Micromeritics). Before the measurement, the samples were degassed at 573 K for 3 h. The X-ray power diffraction (XRD) was carried out on a D/MAX-2600PC diffractometer (Rigaku) operated at 40 kV and 100 mA with Ni-filtered Cu Kα (0.15418 nm) radiation source. The patterns were collected from 10° to 70° (2θ) with a step width of 0.02°. Hydrogen temperature programmed reduction (H₂-TPR) was performed with an Auto Chem II chemisorption analyzer (Micromeritics) using a thermal conductive detector (TCD). The sample was pretreated by He (30 mL/min) at 573 K for 1 h to remove surface contaminants. After cooling to 273 K, 10% H₂/Ar (30 mL/min) mixture was introduced and the temperature was raised to 873 K at a rate of 10 K/min. The dispersion of Pd was measured by CO pulse chemisorption at 273 K. The transmission electron microscopy (TEM) images were obtained on a JEM-2100 (JEOL) instrument at an accelerating voltage of 200 kV.

The CO catalytic oxidation test was performed in a continuous-flow fixed-bed reactor (8 mm internal diameter) at atmospheric pressure. 0.5 g catalyst powder was loaded between two layers of quartz wool. Before the test, the as-prepared Pd/Fe₂O₃ catalysts were reduced by 10% H₂/N₂ (100 mL/min) at 353 K for 1 h. After the catalysts were cooled to 273 K under N₂ (90 mL/min), the feed gas containing 100 ppm CO balanced with compressed air was passed through the reactor at 1000 mL/min. The reaction temperature, measured with a thermocouple, was increased from 273 K to 333 K at intervals of 10 K. CO concentration was

monitored by an online CO analyzer Model 48i (Thermal Scientific).

The CO conversion was calculated according to the following equation,

$$X(\%) = (\text{CO}_{inlet} - \text{CO}_{outlet}) / \text{CO}_{inlet} \times 100$$

where *X* is the CO conversion, CO_{inlet} is the initial CO concentration in the inlet, and CO_{outlet} is the CO concentration in the outlet.

Results and discussion

Characterizations of the as-prepared catalysts

The ICP results of Pd content and BET surface areas of the as-prepared Pd/Fe₂O₃ catalysts are listed in Table 1. The Pd content was in the range of 1.82-1.89 wt%, closing to the theoretical value of 2.0 wt%. The BET specific surface area of the as-prepared Pd/Fe₂O₃-573 catalyst was 180 m²/g, whereas, it was decreased sharply to 79 and 35 m²/g for Pd/Fe₂O₃-673 and Pd/Fe₂O₃-773 catalyst, respectively. These results demonstrated that calcination at high temperature could dramatically decrease the BET specific surface of the as-prepared catalysts. This might be associated with that the number of support pores were decreased or the supports pores were blocked by the entry of some Pd species at higher temperatures [17].

Table 1 The chemical and physical properties of the Pd/Fe₂O₃ catalysts

Samples	BET (m ² /g)	Pd loading (wt%)	Pd dispersion (%)	Fe ₂ O ₃ size (nm)	Pd size (nm)
Pd/Fe ₂ O ₃ -573	180	1.87	55	9 ^a 5-10 ^b	1.8
Pd/Fe ₂ O ₃ -673	79	1.89	51	22 ^a 20-25 ^b	2.0
Pd/Fe ₂ O ₃ -773	35	1.82	4.7	31 ^a 30-40 ^b	2.1

Note: ^athe size of Fe₂O₃ calculated from Scherer formula; ^bthe size range of Fe₂O₃ judged from the TEM images; ^cthe Pd size was calculated according to $d_{Pd} = 1/D_{Pd} \times 100\%$

Figure 1 presents the XRD patterns of the as-prepared Pd/Fe₂O₃ catalysts. There is no observable PdO diffraction peak for all the catalysts, indicating that PdO is finely dispersed. The PdO nanoparticles size was maintained at about 2 nm in diameter (calculated according to the dispersion of Pd, Table 1) even after calcination at 773 K. While for the Fe₂O₃ support, the main diffraction peaks are all assigned to hexagonal Fe₂O₃ (PDF#33-0664). The support crystalline size was 9 nm for Pd/Fe₂O₃-573 catalyst, and it was

increased to 22 nm for Pd/Fe₂O₃-673 and 31 nm for Pd/Fe₂O₃-773 catalysts with the calcination temperature increasing, which is in accordance with the BET specific surface results.

The sintering of the Fe₂O₃ support is further confirmed by the TEM images shown in Fig. 2. The Pd/

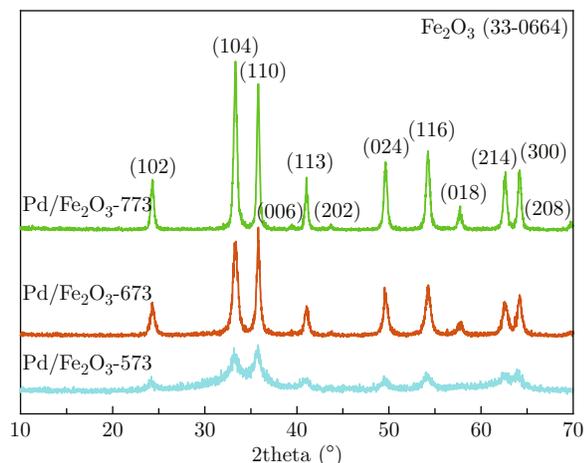


Fig. 1 XRD patterns of the as-prepared Pd/Fe₂O₃ catalysts.

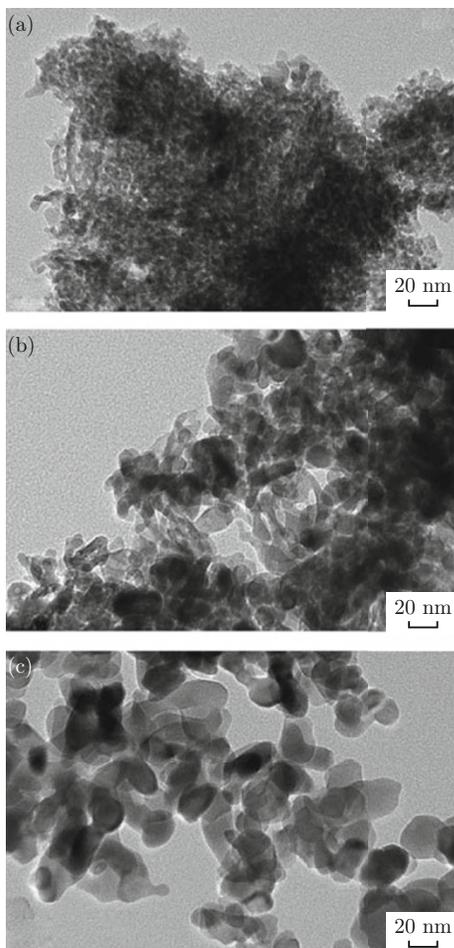


Fig. 2 TEM images of the as-prepared Pd/Fe₂O₃ catalysts: (a) 573 K; (b) 673 K; and (c) 773 K.

Fe₂O₃-573 catalyst had the average Fe₂O₃ crystalline size of ~10 nm (see Fig. 2(a)). When the calcination temperature was increased to 673 K (Pd/Fe₂O₃-673), the Fe₂O₃ size was increased to ~20 nm (see Fig. 2(b)), and further increased to ~30 nm at 773 K (Pd/Fe₂O₃-773) (see Fig. 2(c)). There is no visible PdO nanoparticle, because the 2 nm PdO particle is impossible to be distinguished from the speckle contrast exhibited by the support.

H₂-TPR experiments were carried out on the as-prepared Pd/Fe₂O₃ catalysts, and the profiles are displayed in Figure 3. The temperatures of reduction peaks maxima and the quantitative evaluation of H₂ consumption are given in Table 2. The reduction peaks of PdO species were centered at 316~340 K [14,21]. Among these three catalysts, Pd/Fe₂O₃-573 exhibited the largest H₂ consumption below 400 K (shown in Table 2), which might be associated with the strongest metal-support interaction [22]. In the case of Pd/Fe₂O₃-573 catalyst, the H₂ consumption at 340 K was 355 μmol/g_{cat}, which was much larger than 155 μmol/g_{cat} of theoretical value for PdO to Pd, indicating the co-reduction of PdO and partial surface Fe₂O₃ near the Pd nanoparticles where hydrogen can easily spillover onto the support surface [23]. The reduction peak centered at 450 K (H₂ consumption is 352 μmol/g_{cat}) was due to the further reduction of Fe₂O₃ to Fe₃O₄. It appears that Pd could enhance the Fe₂O₃ support reducibility significantly since the reduction temperature was much lower than the typical one of 633 K for Fe₂O₃ to Fe₃O₄ [24,25]. The reduction behaviors of Pd/Fe₂O₃-673 and Pd/Fe₂O₃-773 catalysts were similar to that of the Pd/Fe₂O₃-573 catalyst. However, the Pd enhancement becomes weaker by considering that the H₂ consumption at low temperature (~320 K) was decreased and the reduction temperature for Fe₂O₃ to Fe₃O₄ was shifted to higher temperatures.

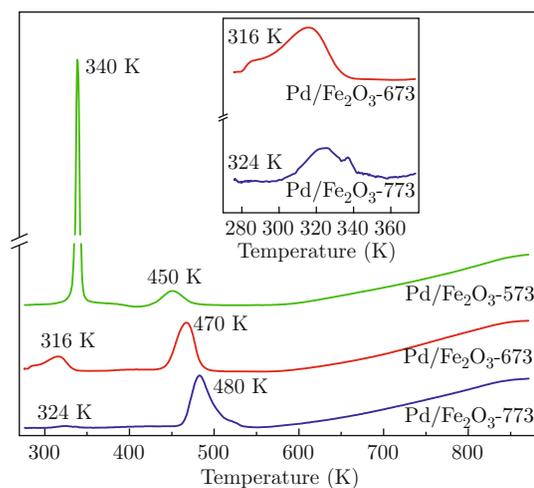


Fig. 3 H₂-TPR profiles of the as-prepared Pd/Fe₂O₃ catalysts.

Table 2 H₂-TPR analysis of the Pd/Fe₂O₃ catalysts

Samples	Reduction Temperature (K)	Hydrogen Consumption (μmol/g)	Contributions
Pd/Fe ₂ O ₃ -573	340	355	PdO → Pd; Fe ³⁺ → Fe ²⁺
	450	352	Fe ³⁺ → Fe ²⁺
Pd/Fe ₂ O ₃ -673	316	327	PdO → Pd; Fe ³⁺ → Fe ²⁺
	470	624	Fe ³⁺ → Fe ²⁺
Pd/Fe ₂ O ₃ -773	324	249	PdO → Pd; Fe ³⁺ → Fe ²⁺
	480	953	Fe ³⁺ → Fe ²⁺

Catalytic performance of the Pd/Fe₂O₃ catalysts in CO oxidation

Catalytic activity of the Pd/Fe₂O₃ catalysts

Figure 4 shows the catalytic activity in CO oxidation of the as-prepared and H₂-pretreated Pd/Fe₂O₃ catalysts. For the as-prepared catalysts, (see Fig. 4(a)) the T_{50} (the temperature corresponding to 50% CO conversion) was 287 K for Pd/Fe₂O₃-573, 309 K for Pd/Fe₂O₃-673, and 313 K for Pd/Fe₂O₃-773 catalysts, respectively. A 100% CO conversion was observed on the Pd/Fe₂O₃-573 catalyst at 333 K, whereas, they were corresponded to only 80% and 70% for the

Pd/Fe₂O₃-673 and Pd/Fe₂O₃-773 catalysts at the same temperature. The results indicated that CO could be oxidized by PdO species without pre-reduction [25]. This might be due to the redox behavior of the highly dispersed PdO nanoparticles: PdO + CO → Pd + CO₂, 2Pd + O₂ → 2PdO.

The H₂ pretreatment promoted the catalytic activity of the Pd/Fe₂O₃ catalysts, and the results are shown in Fig. 4(b). The CO conversion at 273 K was 50~55% for the Pd/Fe₂O₃-773 and Pd/Fe₂O₃-673 catalysts, which was slightly lower than 65% for the Pd/Fe₂O₃-573 catalyst. For all the three catalysts, the CO conversion was about 70% at 283 K, and increased to 100% at 323 K. The much higher performances of the H₂-pretreated Pd/Fe₂O₃ catalysts may be due to the well dispersed Pd nanoparticles and the unique Pd-Fe₂O₃ interaction [26]. It was considered that the well dispersed Pd nanoparticles would weaken C-O and O-O bonds after CO and O₂ adsorption, influencing the interaction between the surface and the adsorbents and favoring CO activation [20,27].

The influence of Pd loading content

The influence of amounts of Pd loading on CO oxidation conversion for the as-prepared and H₂-pretreated Pd/Fe₂O₃ catalysts is shown in Fig. 5. It is clearly seen that the CO conversion was all improved when the loaded Pd content was increased. For the as-prepared Pd/Fe₂O₃-573 catalyst, the CO conversion was increased from 68% to 90% when Pd content was increased from 1 wt% to 3 wt%. Similarly, it was increased from 79% to 99% for H₂-pretreated Pd/Fe₂O₃-573 catalyst. This may be explained by the more active sites which were produced as the loaded Pd content increased for CO adsorption, activation and intermediates transformation.

The influence of gas hourly space velocity (GHSV)

The catalytic performances of the as-prepared and H₂-pretreated Pd/Fe₂O₃-573 catalysts under different gas hourly space velocity (GHSV) were performed at 303 K (Fig. 6). For all the catalysts, a higher space

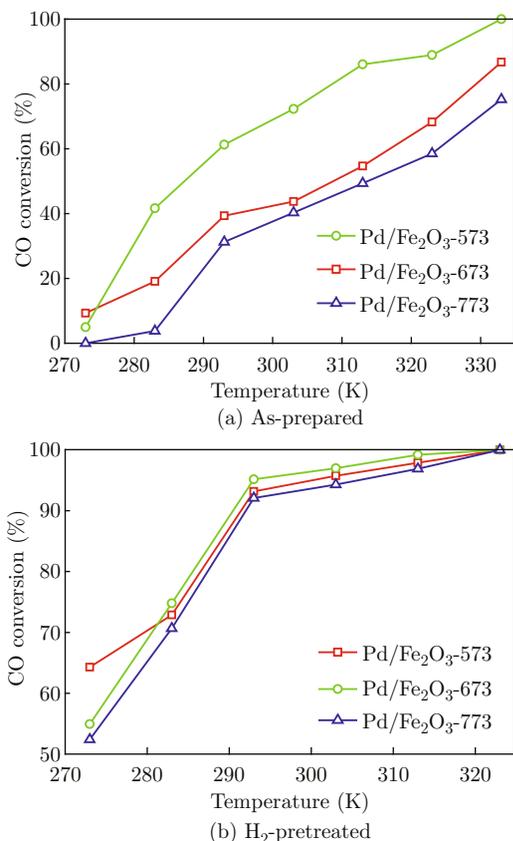


Fig. 4 Catalytic activity in CO oxidation of the Pd/Fe₂O₃ catalysts (Reaction conditions: mass of catalyst: 0.5 g; Total flow rate: 1000 mL/min; Feedstock: 100 ppm CO/air; GHSV=120,000 mL/gh).

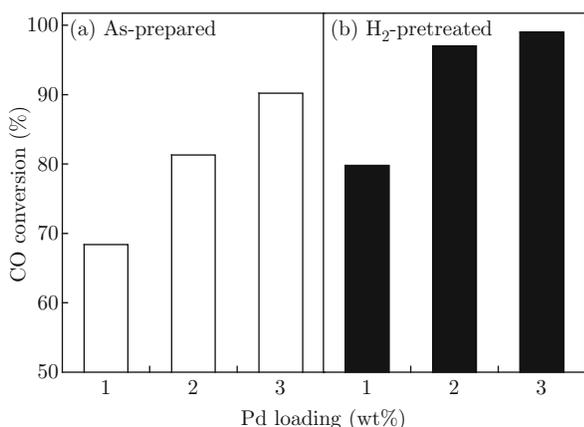


Fig. 5 The influences of Pd loading content on CO conversion over the (a) as-prepared and (b) H₂-pretreated Pd/Fe₂O₃ catalysts calcined at 573 K (Reaction conditions: mass of catalyst: 1.0 g; Total flow rate: 1000 mL/min; Feedstock: 100 ppm CO/air; T: 303 K, GHSV=60000 mL/gh).

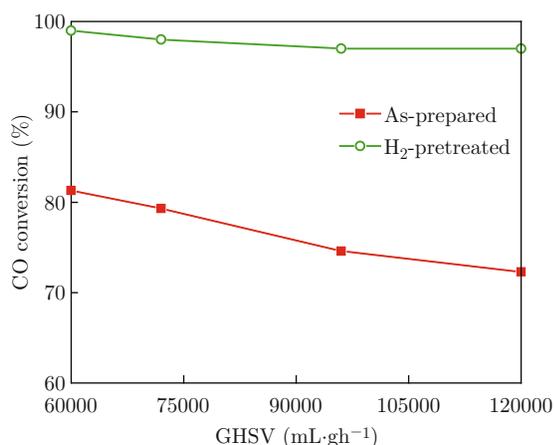


Fig. 6 The Influences of gas hourly space velocity on CO conversion over the as-prepared and H₂-pretreated Pd/Fe₂O₃ catalysts calcined at 573 K (Reaction conditions: mass of catalyst: 0.5~1.0 g; Total flow rate: 1000 mL/min; Feedstock: 100 ppm CO/air; T: 303 K).

velocity resulted in a lower CO conversion. This was accounted on the decreased reactants residence time on the catalyst surface with the increased GHSV. However, the decrease extents were different. The CO conversion

was decreased from 81% to 72% when the space velocity was increased from 60,000 mL/(gh) to 120,000 mL/(gh) over the as-prepared Pd/Fe₂O₃-573 catalyst. While for the H₂-pretreated catalysts, the CO conversion was only slightly decreased from 99% to 97% with the gas hourly space velocity was increased from 60,000 mL/(gh) to 120,000 mL/(gh). The results suggested that the CO oxidation reaction rate was much faster on the H₂-pretreated catalyst than that on the as-prepared catalyst.

The intrinsic rate of CO conversion

The excellent activity of the Pd/Fe₂O₃ catalysts can be further confirmed from Table 3, where a comparison of the specific rate of the as-prepared and H₂-pretreated Pd/Fe₂O₃ catalysts for CO oxidation with a range of supported Pd and Au catalysts from the literatures has been made. The as-prepared Pd/Fe₂O₃ catalyst showed a relative high specific rate (4.3×10^{-2} mol/g_{Pd}h), which was as active as Au/La₂O₃ catalyst (specific rate is 4.0×10^{-2} mol/g_{Au}h). The H₂ pretreatment enhanced the activity further, the specific rate over the H₂-pretreated Pd/Fe₂O₃ catalyst was increased to 10.7×10^{-2} mol/g_{Pd}h, which was a little higher than that of the Au/Fe₂O₃ catalyst (9.1×10^{-2} mol/g_{Au}h) prepared by deposition-precipitation method [17,28]. These results showed that the activities of Fe₂O₃-supported Pd catalysts may be comparable to those of supported Au catalysts for CO oxidation.

In order to have an insight into the intrinsic activities of the Pd catalysts, the turn over frequencies (TOFs) were also compared in Table 3. As expected, the TOF of H₂-pretreated Pd/Fe₂O₃-573 catalyst was 0.038 s^{-1} , exhibiting significantly higher activity than that of the as-prepared Pd/Fe₂O₃-573 catalyst (TOF = 0.012 s^{-1}). The TOF of the H₂-pretreat Pd/Fe₂O₃-573 catalyst was comparable to that of the Au/CeO₂ catalyst (TOF = 0.047 s^{-1}) [29] and the commercial Au/Fe₂O₃ catalyst (TOF = 0.04 s^{-1}) [14]. Taken reaction temperature into account, the H₂-pretreat Pd/Fe₂O₃-573 catalyst is much active than the Pd/SiO₂ catalyst (298 K vs. 450 K, TOF= 3.1 s^{-1}) [30]. The TOF value of H₂-pretreat

Table 3 Specific rates of CO conversion and TOFs data of catalysts from this work and literatures

Catalysts	Metal loadings (wt%)	Specific rate $\times 10^2$ (mol _{CO} · g _{Pd/Au} ⁻¹ · h ⁻¹)	TOF $\times 10^2$ (s ⁻¹)	Temperature (K)	References
As-prepared Pd/Fe ₂ O ₃ -573 ^a	1.87	4.3	1.2	298	This work
H ₂ -pretreated Pd/Fe ₂ O ₃ -573 ^a	1.87	10.7	3.8	298	This work
Pd/Fe ₂ O ₃ ^a	2.1	1.4	3.7	300	[17]
Pd/SiO ₂ ^b	5.0	NA ^c	3.1	450	[31]
Au/Fe ₂ O ₃ ^b	4.4	9.1	4.0	300	[17]
Au/La ₂ O ₃ ^b	NA ^c	4.0	1.0	298	[29]
Au/CeO ₂ ^b	5.0	NA ^c	4.7	273	[30]

Note: ^aCo-precipitation; ^bDeposition-precipitation; NA: Not available

Pd/Fe₂O₃-573 catalyst was in great agreement with the same Pd/Fe₂O₃ catalyst reported in reference [17]. This was assigned to the small size of the synthesized Pd nanoparticles (<3 nm), suggesting much smaller activation energy [31]. It was well known that the size of metal particles plays an important role in CO activation. The TOF results strongly suggested that supported Pd catalysts may be a good alternative to gold in CO oxidation if proper preparation method and support are adopted.

Stability of the Pd/Fe₂O₃ catalysts for CO oxidation

The stability of Pd/Fe₂O₃ catalysts shown in Fig. 7 was tested at 293 K. The CO conversion was readily decreased from 70% to 65% for the Pd/Fe₂O₃-573 catalyst after 10 h (Fig. 7(a)). For Pd/Fe₂O₃-673 catalyst it maintained around 40%, whereas, for Pd/Fe₂O₃-773 catalyst it decreased from 35% to 25% after 10 h. The stability test further evidenced the highest performance of the as-prepared Pd/Fe₂O₃-573 catalyst. Because the reaction temperature (293 K) was much lower than the

calcination (573 K) and reduction temperature (353 K), it was inferred that no obvious PdO or Fe₂O₃ sintering was occurred during the reaction course. Hence the deactivation of the as-prepared Pd/Fe₂O₃ catalysts might be originated from the gradually deposition of soft carbonate or carbonyl species on the catalyst surface.

Figure 7(b) shows the stability of CO conversion over the H₂-pretreated Pd/Fe₂O₃ catalysts. The initial CO conversion was about 90%, and the CO conversion was decreased with the time on stream increasing over the three catalysts. After 10 h, CO conversion was decreased to 80%, 70% and 40% over the Pd/Fe₂O₃-573, Pd/Fe₂O₃-673 and Pd/Fe₂O₃-773 catalysts, respectively. The results demonstrated that Pd/Fe₂O₃-573 catalyst is the most efficient for CO oxidation. It might be associated with the smallest size of Pd (1.8 nm) and the strongest metal-support interaction (Fig. 3) over the Pd/Fe₂O₃-573 catalyst. Although the Pd sizes over the Pd/Fe₂O₃-673 and Pd/Fe₂O₃-773 catalysts were close (2.0 nm and 2.1 nm, respectively), the metal-support interaction was not as strong as that over the Pd/Fe₂O₃-573 catalyst. Similar to the as-prepared catalysts, the deactivation of the H₂-pretreated catalysts could also be contributed to the gradually deposition of carbonates on the surface of catalyst, which hindered the activation of CO and blocked the transformation of intermediates, as discussed later.

Characterizations of the used catalysts

XRD patterns

In order to clarify the causes of deactivation of the catalysts, characterizations of the used catalysts were carried on. Figure 8 gives the XRD patterns of the used as-prepared and H₂-pretreated Pd/Fe₂O₃ catalysts. The XRD patterns of the used as-prepared Pd/Fe₂O₃ catalysts (Fig. 8(a)) showed the exact same profiles with the fresh ones. The Fe₂O₃ diffraction peaks were present and their crystalline sizes kept constant. The diffraction peak of PdO species was still not detected. These observations confirmed that the sintering of PdO and Fe₂O₃ species were not occurred. In contrast, obvious differences were observed over the used H₂-pretreated Pd/Fe₂O₃ catalysts (Fig. 8(b)). The diffraction peaks of Fe₂O₃ species were much weaker and the diffraction peaks of Fe₃O₄ species were appeared. This indicated that the hydrogen pretreatment reduced most of the Fe₂O₃ to Fe₃O₄. Pd species were not detected in these used samples, indicating that they were highly dispersed or below the limitation of XRD apparatus. Hence Pd/Fe₃O₄ catalysts were formed after the hydrogen pretreatment. It has been reported that Fe₃O₄ contained surface hydroxyl group and was helpful for oxygen activation [17]. So the catalytic performance of the H₂-pretreated catalysts was much higher than that

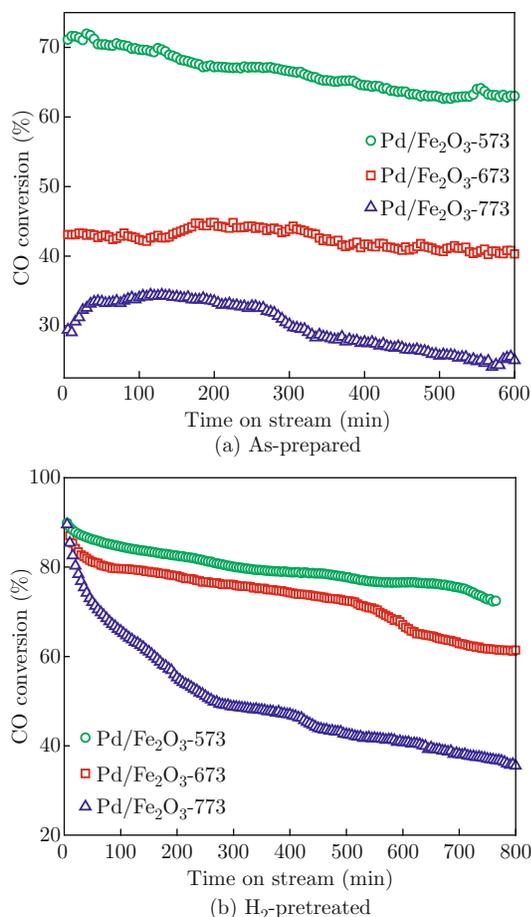


Fig. 7 CO conversion of the Pd/Fe₂O₃ catalysts as a function of time at 293 K (Reaction conditions: mass of catalyst: 0.5 g; Total flow rate: 1000 mL/min; Feedstock: 100 ppm CO/air; T: 293 K; GHSV=120,000 mL/gh).

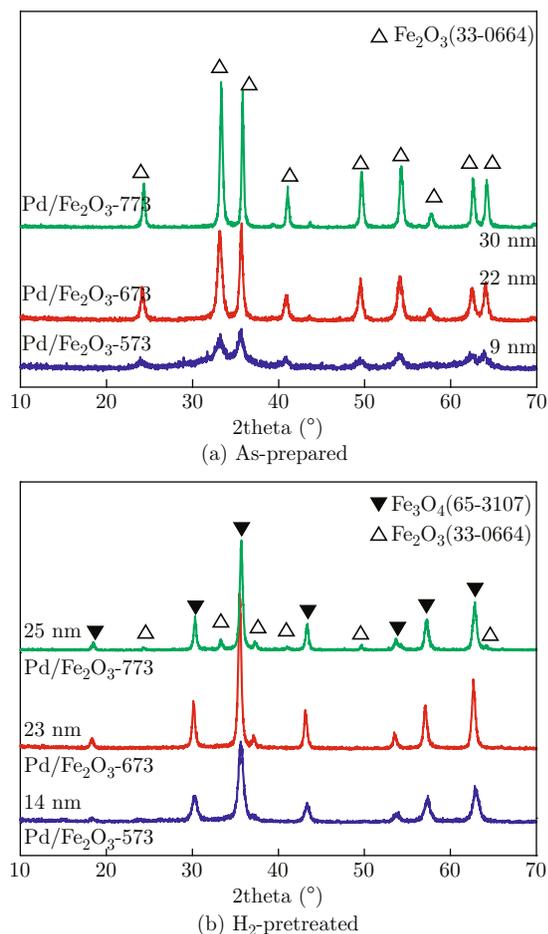


Fig. 8 XRD patterns of used (a) as-prepared and (b) H₂-pretreated Pd/Fe₂O₃ catalysts after the stability test.

of the as-prepared catalysts.

FTIR analysis of the H₂-pretreated Pd/Fe₂O₃ catalysts

Figure 9 presents the FTIR analysis of the H₂-pretreated Pd/Fe₂O₃ catalysts to identify the carboxyl species on the catalyst surface. In the carbonate-like species range (1000-1700 cm⁻¹) [32], the peak at 1415 cm⁻¹ was related to the unidentate carbonate species [33], and 1539 cm⁻¹ was detected for the carboxylate species [13]. The band observed at 1630 cm⁻¹ was assigned to the bidentate carbonates [9,34]. In the carbonyl region (1800-2250 cm⁻¹), CO adsorption and activation was observed. 1977 cm⁻¹ was due to CO bridged adsorption on Pd⁰ and 2162 cm⁻¹ was related to the CO interacting with partially oxidized Pd center [13]. The IR spectroscopy clearly demonstrated that the carbonate species were formed after the stability tests and gave an insight on the activation of CO. It was suggested that CO was adsorbed on the Pd⁰ and activated on the partially oxidized Pd (because of the presence of oxygen in the feed). The intermediates (carbonates or carbonyl species) were transformed to CO₂

in the Pd-Fe₂O₃ interface. However, due to the much less adsorption and activation of CO, the catalytic performance of the as-prepared catalysts was not as good as that of the H₂-pretreated catalysts. In addition, the carbonates on the surface would further cover the surface of the active sites and block the mobility and transformation of the intermediates, resulting in the deactivation with the time on stream [35].

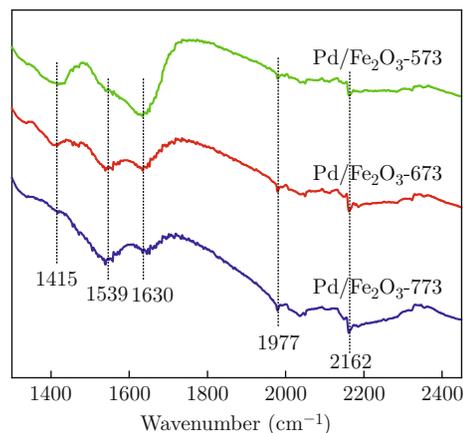


Fig. 9 IR analysis of the used H₂-pretreated Pd/Fe₂O₃ catalysts after the stability test.

Conclusions

Pd/Fe₂O₃ nano-catalysts were synthesized by co-precipitation method and were investigated for CO oxidation in this study. The results revealed that both the as-prepared and H₂-pretreated catalysts showed high performance for CO catalytic oxidation at low temperatures. T₅₀ was 293 K and 273 K over the as-prepared and the H₂-pretreated catalysts, respectively. CO can fully be converted at temperature as low as 313 K over the H₂-pretreated catalysts. During the long time stability test, CO conversion was above 80% over the H₂-pretreated Pd/Fe₂O₃-573 catalyst, while 60% conversion was maintained over the as-prepared Pd/Fe₂O₃-573 catalyst. The higher calcination temperature deduced a lower performance, which was associated with the different crystalline size and metal-support interaction of the catalyst. The much higher performance of the H₂-pretreated catalysts was attributed to the Fe₃O₄ formation and surface OH group. The former was helpful to the O activation, and the latter was reacted with CO to form reactive adsorbed intermediates. The deactivation caused of the catalyst was assigned to the carbonates formation on the surface that hindered the CO activation and intermediates transformation.

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