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ARTICLE

Synthesis of Sr-modified Pd-Rh Bimetallic Catalyst with Advanced Three-Way Catalytic Performance

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Abstract: A series of SrO-modified Pd-Rh/Al₂O₃ catalysts were synthesized by co-impregnation technique, i. e., Sr precursor was introduced into the mixed salt solution of Pd and Rh prior to their impregnation on Al₂O₃ powders. The results reveal that owing to the diffusion barrier effect of SrO, the introduction of appropriate amount of SrO (1wt%~2wt%) can increase the dispersion of Pd and Rh, as well as improve the hydrothermal stability of the catalyst. After hydrothermal aging treatment, higher dispersion and more active oxidation state of Pd and Rh are maintained. Consequently, modified reduction capability and improved three-way catalytic performance can be obtained for the appropriate SrO-modified Pd-Rh/Al₂O₃ catalyst. However, when excessive amount of SrO is introduced, plenty of surface sites on Al₂O₃ support will be occupied, and new SrAl₂O₄ phase is generated, leading to adverse effect instead. To sum up, the catalyst modified by 2wt% SrO exhibits the best three-way catalytic performance, by which after hydrothermal aging treatment, the light-off temperatures of CO, HC and NO are 23, 15 and 27 °C lower than those by bare Pd-Rh/Al₂O₃ catalyst.

Key words: SrO; Pd-Rh/Al₂O₃ catalysts; dispersion; oxidation state; three-way catalytic performance

Owing to the prominent efficiency to simultaneously eliminate the three main contaminants (CO, HC and NO) in exhaust emissions of gasoline engine vehicles, installation of three-way catalytic converters has become an universally applied approach to control emissions in exhaust gas aftertreatment industry^[1-4]. Generally speaking, three-way catalysts (TWCs) are usually composed of platinum group metals (PGMs) dispersed in support materials (such as high-surface-area Al₂O₃ material)^[5-9]. In practical application of TWCs, Pd is widely used as an active component owing to their outstanding oxidation activity to HC/CO and prominent high-temperature aging resistance, while Rh is often also indispensable on account of its excellent NO purification performance^[10]. Pd-Rh bimetallic catalyst has been demonstrated to be currently the most commonly applied

commercial TWC system^[7,11,12]. In recent years, persistent increase of the price of Pd and Rh brings about more and more cost pressure. Therefore, to develop Pd-Rh catalysts with improved three-way catalytic performance by promoting the high dispersion and modifying the oxidation state of noble metals in the support materials is in urgent demand, so that the usage amount of Pd and Rh can be befittingly lowered for cost reduction.

It is well known that the introduction of rare earth and alkaline earth metal oxides, such as La₂O₃, CeO₂, Pr₆O₁₁, BaO, SrO, CaO, into the supported noble metal catalyst systems, has been reported by many researchers to have the ability to promote the dispersion and to modify the chemical state of the active components, as well as to enhance the thermal stability of the support materials and the catalysts, which consequently

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contributes to their improved catalytic performance^[13-22]. In addition, the additives may also act as other electron promoter or NO_x adsorbent to effectively promote the NO_x purification performance^[13,23-25]. In practical application, SrO has been applied as auxiliaries in TWC, while the detailed influence of SrO on the properties of the catalysts has seldom been reported. In addition, the aforementioned additives are most commonly introduced during the preparation of the support materials, or introduced into the catalyst system prior to the impregnation of noble metals, or introduced into the as-prepared noble metal catalyst. In this work, SrO-modified Pd-Rh/Al₂O₃ catalyst was synthesized by co-impregnation of appropriate amount of Sr and Pd, Rh precursors on Al₂O₃ support, and the influence of SrO on the physicochemical properties of the catalyst was investigated, so that Pd-Rh bimetallic catalyst with advanced three-way catalytic performance can be achieved.

1 Experiment

1.1 Synthesis of Pd-Rh bimetallic three-way catalysts

A Pd-Rh/Al₂O₃ catalyst without the participation of SrO was prepared by a co-impregnation method. The γ -Al₂O₃ powders were supplied by Solvay Company. The precursors of Pd and Rh, i.e., Pd(NO₃)₂ solution and Rh(NO₃)₃ solution, were both purchased from Shanghai Jiuyue Chemical Co., Ltd, and the concentrations of Pd and Rh in the salt solutions were 8.47wt% and 15.00wt%, respectively. In addition, the loading contents of Pd and Rh on Al₂O₃ were controlled at 2wt% and 1wt%, respectively. The detailed synthesis procedure was as follows: firstly, 2.36 g of Pd(NO₃)₂ solution and 0.67 g of Rh(NO₃)₃ solution were placed in a glass beaker; after blended thoroughly, 10 g of γ -Al₂O₃ powders were introduced into the mixed solution, and evenly stirred for 20 min. The impregnated mixture was then submitted to drying at 100 °C for 3 h and calcined at 550 °C for 3 h to get the catalyst powders. Afterwards, the catalyst powders were introduced into distilled water and mixed thoroughly, and then a slurry was obtained, which was subsequently coated onto a cordierite monolith (400 cells/in², Corning, USA), followed by drying at 100 °C for 3 h and calcining at 550 °C for 3 h, and hence the monolithic Pd-Rh/Al₂O₃ catalyst was eventually obtained, hereafter labeled as PdRh.

A series of SrO-modified Pd-Rh/Al₂O₃ catalysts were synthesized by almost the same procedure as described above, except that during the preparation of mixed solution of Pd and Rh, required amount of Sr(NO₃)₂ aqueous solution was introduced and blended together, i.e., a mixed solution of Pd, Rh and Sr was impregnated onto the γ -Al₂O₃ powders. Certainly, the impregnation, drying and calcining processes, along with the coating process, were all the same as those for bare PdRh catalyst. And the obtained catalysts with SrO content of 1wt%, 2wt%, 4wt%, 8wt% were named as Sr1, Sr2, Sr4 and Sr8, respectively.

Moreover, to estimate the hydrothermal aging resistance, the as-prepared five catalysts were further submitted to

treating in an atmosphere composed of 10vol% H₂O/air at 1000 °C for 5 h, and the obtained aged catalysts were correspondingly marked as PdRha, Sr1a, Sr2a, Sr4a and Sr8a.

1.2 Characterization of the catalysts

The surface areas and pore volumes of the fresh and aged catalysts were measured by an Autosorb SI Quantachrome automated surface area and pore size analyzer. The samples were firstly degassed under vacuum at 300 °C for 3 h and then underwent a nitrogen adsorption-desorption procedure at -196 °C.

Powder X-ray diffraction (XRD) patterns were collected using a Rigaku DX-2500 diffractometer. Cu K α (λ =0.154 nm) was employed as the radiation source. The XRD patterns in the 2 θ range of 10°~90° were collected with a step size of 0.02°.

Raman characterization was performed employing a Renishaw inVia reflex Raman spectrometer. An excitation wavelength of 532 nm was applied during the whole testing process. The Raman spectra in the frequency range of 100~1000 cm⁻¹ were recorded with a resolution of 1 cm⁻¹.

X-ray photoelectron spectroscopy (XPS) analysis was conducted on a British Kratos XSAM-800 electron spectroscopy to investigate the surface of the various catalysts. Monochromatic Al K α (1486.6 eV) was chosen as the radiation. The measured binding energies were calibrated using C-1s (284.8 eV) as the internal reference standard.

The transmission electron microscopy (TEM) observation was carried out employing a ZEISS Libra 200 FE transmission electron microscope. Both TEM and HRTEM (high-resolution TEM) images were observed with an accelerating voltage of 200 kV during the experimental process.

Temperature-programmed reduction in H₂ was conducted to evaluate the reduction features of the catalysts. Prior to the reduction process, 0.1 g of catalyst powder was placed in a U-type quartz tube, and N₂ was passed at 400 °C for 1 h. After that the sample was cooled down to room temperature, and the atmosphere was switched to a mixture of 5vol% H₂/N₂. The reduction was then proceeded by raising the temperature at a constant heating rate of 5 °C/min, and the H₂ consumption was continuously monitored by a thermal conductivity detector (TCD).

1.3 Measurement of three-way catalytic performance

The three-way catalytic activity was measured in a fixed-bed continuous flow reactor. The monolithic catalyst was placed at a specific region in the reactor with stable temperature. The temperature was adjusted by a furnace equipped with a temperature controller. A gas mixture which simulated the actual exhaust gas emission from gasoline engine vehicles was passed through. And the gas composition was controlled as follows: 120 μ L/L of C₃H₈, 240 μ L/L of C₃H₆, 5600 μ L/L of CO, 1900 μ L/L of H₂, 1000 μ L/L of NO, 12vol% CO₂, and 10vol% H₂O, O₂ (adjustable) and N₂ as the balance. Before the measurement, the catalyst was pretreated under reactive gas mixture at 550 °C for 1 h, and the concentration of O₂ was adjusted to an appropriate value to ensure a

stoichiometric air-to-fuel ratio condition. The light-off performance of CO, HC and NO was evaluated by monitoring the compositions of the inlet and outlet gas mixtures via Antaris IGS-6700 FTIR (Thermo Fisher Scientific).

2 Results and Discussion

2.1 Surface areas and pore volumes

The specific surface areas (S_{BET}) and pore volumes of the fresh and aged catalysts are displayed in Fig. 1. As can be seen therein, for the fresh samples, the introduction of SrO into the PdRh catalyst results in somewhat lower surface areas and pore volumes, especially for the samples with a comparatively high content of SrO, which may be due to the fact that excessive SrO will occupy a large amount of the surface of Al_2O_3 support. However, it is also noticed that the introduction of appropriate amount of SrO seems to be beneficial to the hydrothermal stability of the catalyst, and the surface areas and pore volumes of Sr1a and Sr2a are a bit larger than those of bare PdRha. Whereas excessive amount of SrO presents inversely negative influence on the textural properties of the catalysts.

2.2 XRD characterization results

The XRD patterns of the fresh and aged catalysts are displayed in Fig. 2. Only two phases, $\gamma\text{-Al}_2\text{O}_3$ and PdO can be identified from Fig. 2a, and no obvious differences can be found for the fresh samples, indicating that the introduction of SrO exerts little influence on the phase of the PdRh catalyst. However, after hydrothermal aging treatment, significant

differences can be found from Fig. 2b. Phase transformation of $\gamma\text{-Al}_2\text{O}_3$ to transitional $\theta\text{-Al}_2\text{O}_3$ takes place during the hydrothermal aging process, and a new phase, SrAl_2O_4 appears when the content of SrO is above 4wt%. Under this condition, the newly formed SrAl_2O_4 may be distributed on the surface of Al_2O_3 , which consequently brings about blockage of the pores, and is in good agreement with the obviously declined surface areas and pore volumes. In addition, with regard to the noble metal species, besides for PdO, characteristic peaks of metallic Pd are also observed, especially for Sr8a, demonstrating that the introduction of excessive amount of SrO is detrimental to the maintenance of high dispersion and oxidative state of Pd species.

2.3 Raman characterization results

Fig. 3 presents the Raman spectra of the fresh and aged catalysts. Therein the most intense band located at 648 cm^{-1} is recognized as PdO phase, which also produces plenty of weak peaks in the frequency range of $100\text{--}1000\text{ cm}^{-1}$ [26-28]. It is generally considered that the position and intensity of the principal peak of PdO are associated with the oxygen state, particle size and concentration of PdO species in the catalysts. For the fresh samples, it is found from Fig. 3a that when the content of SrO is smaller than 4wt%, the intensity of the PdO peak at 648 cm^{-1} for the SrO-modified samples is similar to that of bare PdRh, while Sr8 exhibits an obviously weaker peak at 648 cm^{-1} , indicating that Sr8 possesses obviously smaller amount of PdO species, which is disadvantageous to the three-way catalytic activity. After hydrothermal aging treatment, remarkable peak sharpening can be observed for all samples, which is indicative of particle aggregation of PdO species to large extent. However, it should be pointed out that although the particle sizes of the noble metal species for Sr1a and Sr2a are smaller than that for bare PdRha (as confirmed by the following TEM observation results), the principal PdO peak shows similar intensities for the three samples, implying that more PdO species are present in Sr1a and Sr2a. Whereas Sr4a and Sr8a possess lower content of PdO species. That is, the introduction of appropriate amount of SrO can help Pd species to stay at a more active PdO state, which is consequently beneficial to the three-way catalytic performance.

2.4 XPS analysis results

The surface concentrations of Pd and Rh derived from XPS

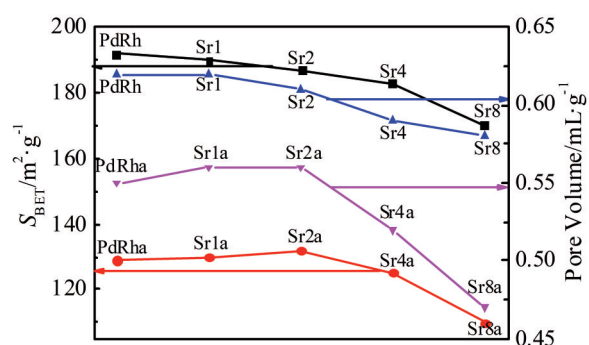


Fig. 1 Specific surface areas (S_{BET}) and pore volumes of the fresh and aged catalysts

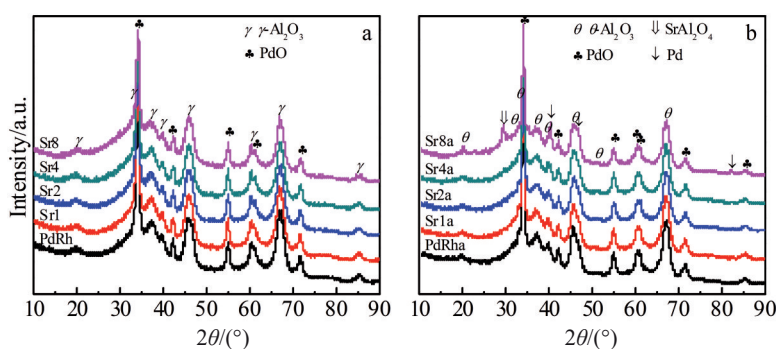


Fig. 2 XRD patterns of the fresh (a) and aged (b) catalysts

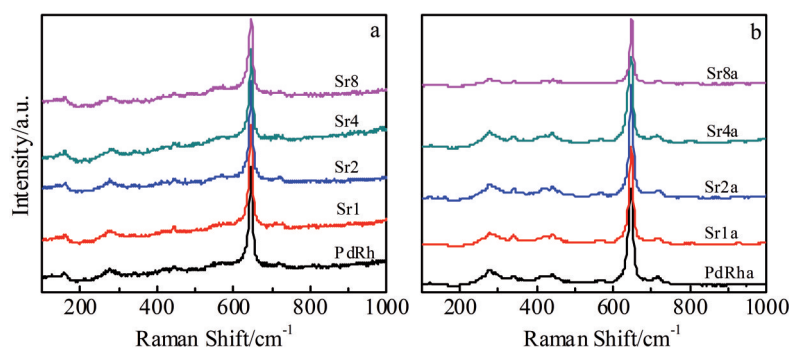


Fig.3 Raman spectra of the fresh (a) and aged (b) catalysts

analysis are illustrated in Fig. 4. As can be seen therein, compared with bare PdRh, Sr1 and Sr2 display higher surface concentrations of Pd and Rh. In other words, introduction of a small amount of SrO into the PdRh catalyst can facilitate the dispersion of noble metal species on the surface. However, when the content of SrO further increases, the surface concentrations of Pd and Rh gradually decrease. After hydrothermal aging treatment, the surface concentration of Pd obviously decreases to a large extent for all catalysts, indicating that severe aggregation of Pd species occurs. Notwithstanding, it is also worth noting that the surface concentration of Rh for the aged catalysts appears to be similar to that for the fresh counterparts, which can be interpreted by the fact that although the catalysts undergo aggregation of Rh species as well, Rh species have larger tendency to locate at the outer surface of the catalysts. In addition, it can be also deduced from Fig.4 that introduction of appropriate amount of SrO into the PdRh catalyst can help to maintain higher surface concentrations of Pd and Rh, and on the whole, Sr2a behaves the best among various aged catalysts.

To better clarify the chemical state of Pd and Rh species, the Pd 3d and Rh 3d XPS spectra are displayed in Fig.5. It can be seen that the Pd 3d curves can be fitted into four peaks, while the Rh 3d curves are generally deconvoluted into six peaks, upon which the Pd²⁺ and Rh³⁺ fractions can be calculated based on the peak areas of different Pd and Rh

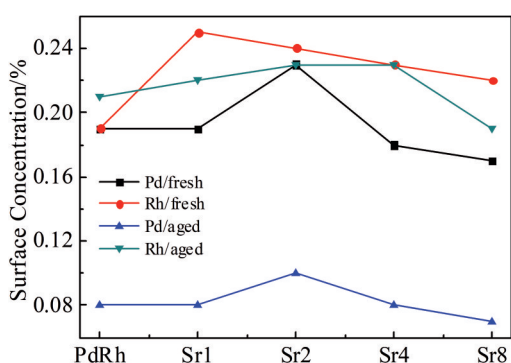


Fig.4 Surface concentrations of Pd and Rh for the fresh and aged catalysts

species. And the obtained results are illustrated in Fig. 6, wherein it is found that the five fresh catalysts possess similar Pd²⁺ fractions, while an increasing tendency of the Rh³⁺ fraction is observed when the content of SrO gradually increases, indicating that the introduction of SrO can help more Rh species to stay at oxidative Rh³⁺ state. After hydrothermal aging treatment, it is found that as the content of SrO increases, both the Pd²⁺ fraction and the Rh³⁺ fraction show a tendency of firstly increasing and then decreasing. That is, the introduction of appropriate amount of SrO into the PdRh catalyst is beneficial to maintain more active oxidation states of Pd and Rh species during hydrothermal aging treatment, consequently contributing well to the three-way catalytic performance. Whereas excessive amount of SrO will inversely produce adverse effect.

2.5 TEM and HRTEM observations

Fig. 7 and Fig. 8 present TEM and HRTEM images of the five aged catalysts respectively. It is observed from Fig.7 that the rod-like particles are identified as Al₂O₃, and the dark particles distributed on Al₂O₃ are mainly noble metal species. It is worth noting that Sr1a and Sr2a possess smaller particle sizes of noble metal species, while Sr4a and Sr8a inversely show increasing particle size of noble metal species. In other words, introduction of appropriate amount of SrO can help maintain relatively higher dispersion of Pd and Rh on Al₂O₃ upon hydrothermal aging treatment, which may be ascribed to the diffusion barrier effect of the SrO additive. In addition, when more SrO is introduced, it may occupy plenty of surface sites on Al₂O₃, which is contrarily disadvantageous to the dispersion of Pd and Rh.

From Fig. 8a₁ and Fig. 8a₂, it can be observed that large particles of metallic Pd and reduction-resistant RhO₂ species are found in PdRha, meaning that during hydrothermal aging process, transformation of Pd and Rh species into more inactive oxidation states has taken place, which will consequently give rise to catalyst deactivation of PdRha to a large extent. Fortunately, when appropriate amount of SrO is introduced, higher hydrothermal stability of the catalyst is achieved. As can be seen in Fig. 8b₁, 8b₂, 8c₁, 8c₂, smaller particle sizes of noble metal species are observed, the majority of which are in more active oxidation states, i.e., PdO and Rh₂O₃. Under this condition, improved three-way catalytic

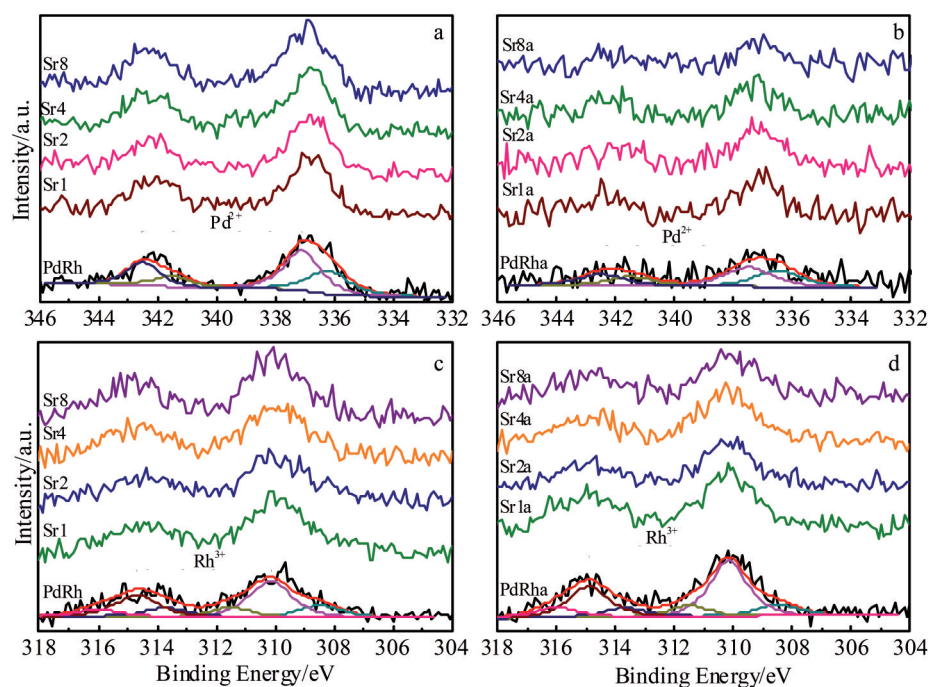


Fig.5 Pd 3d (a, b) and Rh 3d (c, d) XPS spectra of the fresh and aged catalysts

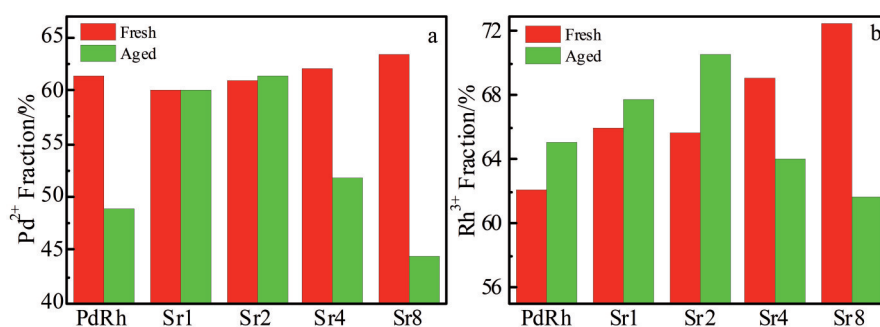


Fig.6 Pd²⁺ fraction (a) and Rh³⁺ fraction (b) of the fresh and aged catalysts

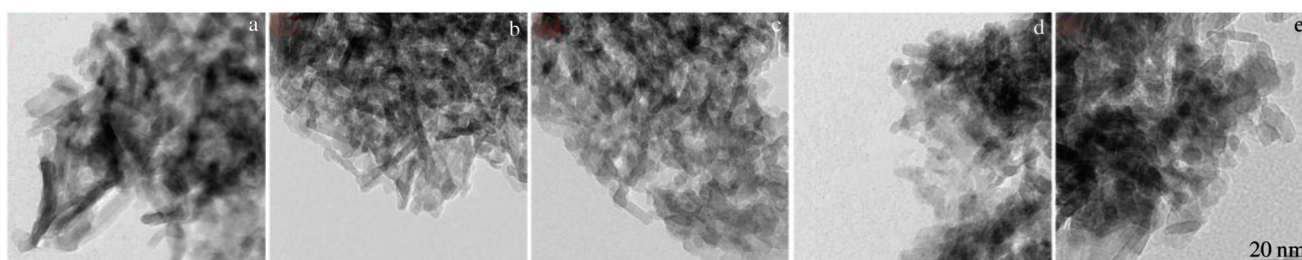


Fig.7 TEM images of PdRha (a), Sr1a (b), Sr2a (c), Sr4a (d) and Sr8a (e)

performance of Sr1a and Sr2a is anticipated. However, when the content of SrO exceeds 4wt%, as mentioned above, a large amount of SrO species may occupy plenty of surface sites on Al₂O₃ support, which is detrimental to the dispersion and hydrothermal stability of the catalyst, that is, noble metal species on Sr4a and Sr8a are more easily to agglomerate and to transform into inactive states.

2.6 H₂-TPR studies

The reduction profiles of the catalysts are depicted in Fig.9.

As can be seen from Fig.9a, the reduction of PdRh occurs at two temperature regions, and the main reduction peak is located at around 50~100 °C, which is attributed to the reduction of PdO species^[29]. In addition, the comparatively weak reduction peak at above 300 °C should be ascribed to the reduction of Rh₂O₃ species on the surface of Al₂O₃^[7,30]. After SrO is introduced into the PdRh catalyst system, the reduction characteristics of the samples with a low content of SrO (Sr1, Sr2) appear to be similar to that of bare PdRh. But when the

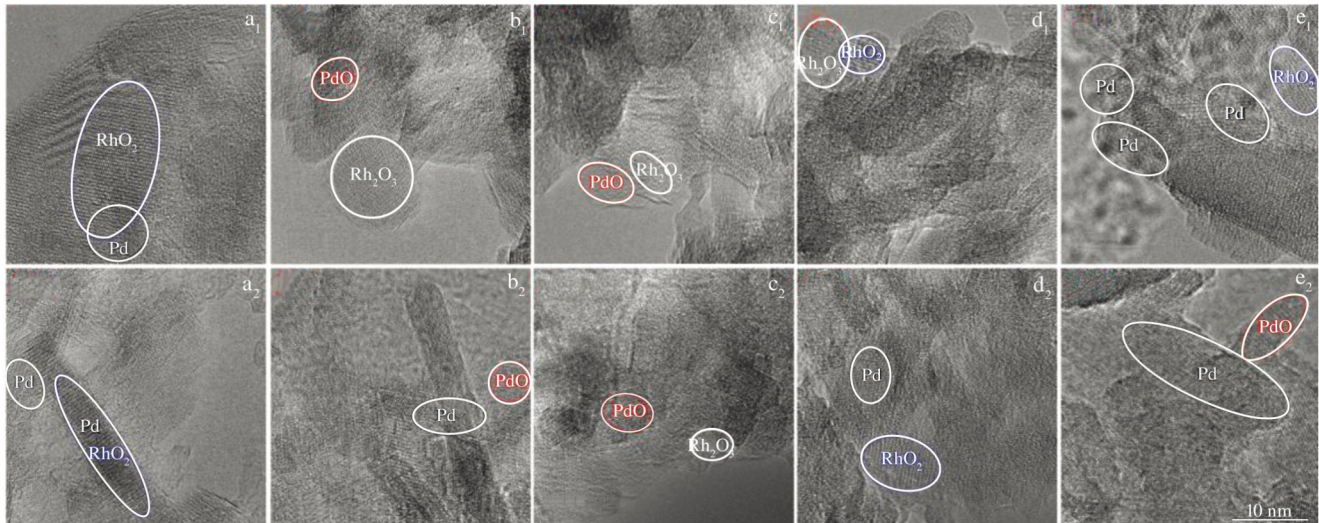


Fig.8 HRTEM images of PdRha (a₁, a₂), Sr1a (b₁, b₂), Sr2a (c₁, c₂), Sr4a (d₁, d₂) and Sr8a (e₁, e₂)

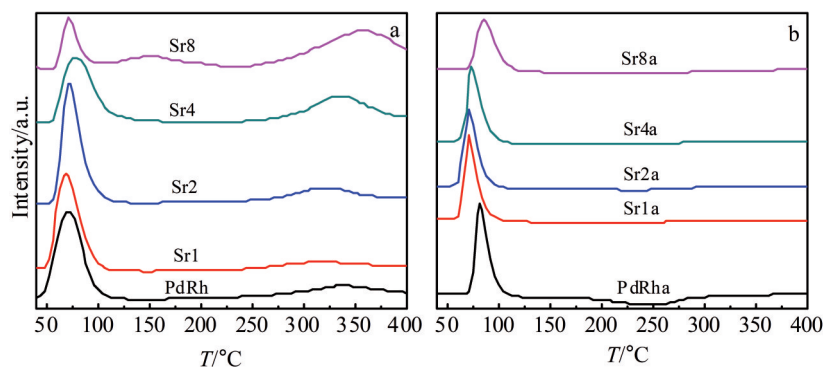


Fig.9 H₂-TPR profiles of the fresh (a) and aged (b) catalysts

content of SrO exceeds 4wt%, the reduction of PdO is obviously weakened. Whereas the reduction of Rh₂O₃ seems to be promoted, implying that the interaction between Rh and Al₂O₃ may be retarded by the additive SrO, which results in the presence of more Rh³⁺ species (as confirmed by the XPS analysis results), and is consequently beneficial to the reduction capability.

After hydrothermal aging treatment, the reduction features of the aged catalysts are intrinsically different from those of their fresh counterparts. As displayed in Fig. 9b, the most obvious difference is that the reduction of Rh₂O₃ species is absent in the experimental temperature range, which is in good consistency with the results of Rh/Al₂O₃ catalyst^[31]. When it comes to the reduction of PdO species, it is found that owing to the severe aggregation and transformation of Pd species to more irreducible state upon hydrothermal aging treatment, the reduction of PdO for bare PdRha obviously shifts towards higher temperature. Fortunately, Sr1a and Sr2a exhibit improved hydrothermal stability, the reduction of which occurs at almost the same temperature as that of fresh Sr1 and Sr2, and the peak intensities are significantly higher compared with those of Sr4a and Sr8a. In summary, among

the five aged catalysts, Sr1a and Sr2a possess the best reduction capability, which is beneficial to their three-way catalytic reaction process.

2.7 Three-way catalytic activity evaluation results

The three-way catalytic activity evaluation results, represented by the light-off temperatures of CO, HC and NO over the fresh and aged catalysts, are presented in Fig. 10. As shown in Fig. 10a, Sr1 and Sr2 exhibit slightly lower light-off temperatures of CO, HC and NO, which can be ascribed to the higher dispersion of Pd and Rh species. However, when comparatively higher content of SrO is introduced, plentiful surface sites of Al₂O₃ will be occupied by excessive SrO, which is disadvantageous to the high dispersion of Pd and Rh, and thus the inferior three-way catalytic performance of Sr4 and Sr8 is reasonable.

A comparison in Fig. 10a and Fig. 10b reveals that the light-off temperatures of CO, HC and NO become obviously higher for the aged catalysts and it can be deduced that hydrothermal aging treatment brings about catalyst deactivation to different extents. As shown in Fig. 10b, as the content of SrO gradually increases, the light-off temperatures of CO, HC and NO firstly decrease and subsequently increase, and on the whole, Sr2a

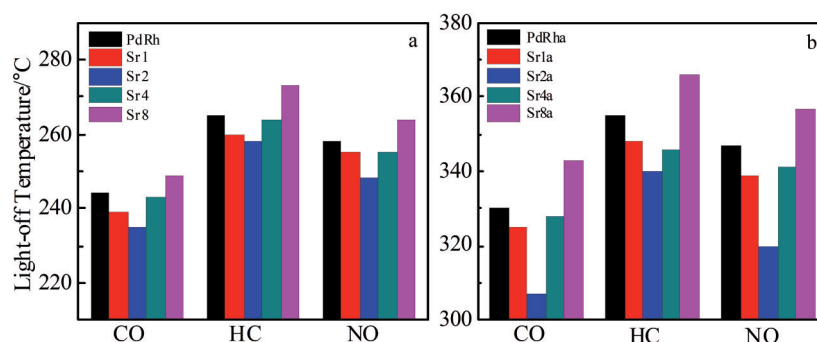


Fig.10 Light-off temperatures of CO, HC and NO for the fresh (a) and aged (b) catalysts

possesses the best three-way catalytic performance. That is, the catalyst modified by appropriate amount of SrO owns the highest hydrothermal stability. The following reasons are considered responsible for this phenomenon. (1) Owing to the diffusion barrier function of SrO, aggregation of noble metal species can be effectively inhibited, and consequently higher dispersion of Pd and Rh species is maintained upon hydrothermal aging process, which can provide more active sites to proceed the three-way catalytic reactions. (2) The introduction of appropriate amount of SrO have the ability of helping Pd and Rh species to stay at more active oxidation states (PdO and Rh₂O₃), which is beneficial to the catalytic performance. (3) The physico-chemical property of SrO is similar to that of BaO, so similar to BaO, SrO may also act as a NO adsorbent. In this way, the contact time between the reaction gas and the catalyst will be prolonged, which can further facilitate the catalytic reaction process.

3 Conclusions

1) Introduction of appropriate amount of SrO can facilitate the high dispersion of Pd and Rh species on Al₂O₃, and enhanced hydrothermal stability can be achieved owing to the diffusion barrier function of SrO. That is, higher dispersion and more favorable oxidation states of Pd and Rh species are maintained for the modified catalyst after hydrothermal aging treatment.

2) SrO may act as a NO adsorbent. Through effective adsorption of NO species, the contact time between the reaction gas and the catalyst can be prolonged, which can also facilitate the catalytic reaction process, and further detailed research is needed to verify this issue.

3) When excessive amount of SrO is introduced, a large amount of SrO may occupy plenty of the surface sites of Al₂O₃, and new phase of SrAl₂O₄ is generated after aging treatment, which is detrimental to the dispersion of Pd and Rh species, and the corresponding catalysts behave undesirable hydrothermal stability. On the whole, after hydrothermal aging treatment, the PdRh/Al₂O₃ catalyst modified by appropriate amount of SrO (2wt%) exhibits excellent reduction capability and superior three-way catalytic performance.

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性能优异的SrO改性Pd-Rh双金属催化剂的制备

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摘 要: 在浸渍到Al₂O₃载体材料上之前, 通过将Sr的前驱体引入到贵金属Pd和Rh的混合盐溶液中, 即采用共浸渍法制备了一系列SrO改性的Pd-Rh/Al₂O₃催化剂。结果表明, 由于SrO的扩散障碍效应, 适量SrO(1%~2%, 质量分数)的引入有利于促进Pd, Rh的分散, 并且能提高催化剂的水热稳定性。水热老化之后, 催化剂能保持更高的Pd, Rh分散度, 并且Pd, Rh能稳定在活性更好的氧化态。因此, 适量SrO改性的Pd-Rh/Al₂O₃催化剂具有更好的还原性能和三效催化活性。然而, 当向催化剂中引入过量的SrO, 由于SrO占据了过量的Al₂O₃的表面, 而且老化后会出现新的物相SrAl₂O₄, 反而会对催化剂的性能产生不利的影响。总的来说, 2% SrO改性的催化剂具有最优异的三效催化活性, 水热老化之后, 相比于空白的Pd-Rh/Al₂O₃催化剂, 2%SrO改性的催化剂使CO, HC和NO的起燃温度分别降低了23, 15和27℃。

关键词: SrO; Pd-Rh/Al₂O₃催化剂; 分散度; 氧化态; 三效催化活性

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