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Communication

Pt nanoparticles encapsulated on V₂O₅ nanosheets carriers as efficient catalysts for promoted aerobic oxidative desulfurization performance

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ABSTRACT

Platinum group metals (PGMs) usually exhibit promising aerobic catalytic abilities, providing a green and feasible oxidative desulfurization method. However, often, the PGM nanoparticles (NPs) get leached, and the catalysts are deactivated. In this work, Pt NPs with particle sizes of approximately 4–5 nm were encapsulated effectively and uniformly on the surface of vanadium pentoxide (V₂O₅) nanosheets (with thicknesses of approximately six atomic layers) through strong metal-support interactions. The synthesized catalysts promote catalytic aerobic oxidation reactions, realizing deep desulfurization (99.1%, < 5 μg g⁻¹) under atmospheric pressure and 110 °C reaction temperature. Remarkable degrees of sulfur removal could be achieved for oils with different initial S-concentrations and substrates. Additionally, the as-prepared catalysts could be recycled for reuse at least seven times.

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Noble metals, especially platinum group metals (PGMs), often exhibit remarkable catalytic abilities in industrial reactions because of their unique electronic *d*-band structures and high valence electron ratios [1–4]. Among their numerous applications, aerobic oxidative desulfurization reactions have received increasing attentions recently, as increasingly restrictive regulations for diesel oils have been introduced. In these reactions, molecular oxygen (O₂) in the air can be activated effectively by synthetic PGM nanoparticles (NPs) [5,6]. Unreactive sulfides

can be subsequently oxidized to polar sulfones and separated out from non-polar oil naturally [7–13]. However, leaching and sintering of PGM NPs often occur during the reaction process, leading to the inevitable deactivation of the catalysts [14]. Thus, supports that could encapsulate PGM NPs effectively and uniformly are desirable.

Among the previously reported supports, redox active transition metal oxides (TMOs), such as ZrO₂, TiO₂, CeO₂ and ZnO, have attracted significant attentions [1–4]. It is principally be-

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Fig. 1. TEM images of Pt NPs-1/V₂O₅ nanosheets (a), Pt NPs-2/V₂O₅ nanosheets (b), Pt NPs-3/V₂O₅ nanosheets (c); (d) HR-TEM image of Pt NPs-2/V₂O₅ nanosheets; (e–f) HR-TEM images of the marked regions in Fig. 1(d).

lied that strong metal-support interactions (SMSIs) would be formed from *d*-orbital overlap between the PGM NPs and the reduced cations of the TMOs [15]. However, the specific surface areas (SSAs) of these redox active TMOs are usually small, making it challenging to disperse PGM NPs onto their surfaces effectively and uniformly. It is known that materials with two-dimensional (2D) morphologies often exhibit larger SSAs

[16]. Thus, redox active TMO carriers with ultra-thin 2D layered structures might be promising candidates. Orthorhombic vanadium pentoxide (V₂O₅), as a redox active TMO, is composed of linked distorted VO₆ octahedra, exhibiting a layered anisotropic structure (Scheme S1) [17]. It is possible to acquire ultra-thin 2D V₂O₅ nanosheets with significantly increased SSAs by relevant exfoliation strategies as the existing van der Waals forces between each layer of the V₂O₅ materials are weak [18]. Additionally, V₂O₅ can readily change its oxidation state between V(VI) and V(V), causing a significant effect on the interactions with dispersed PGM NPs [19].

Herein, Pt NPs were encapsulated on the surface of exfoliated V₂O₅ nanosheets through a convenient sonication-assisted colloidal deposition method [20,21]. The obtained samples are herein referred to as Pt NPs-*n*/V₂O₅ nanosheets. Here, *n* represents the theoretical mass percentage of the Pt NPs in the material, which was confirmed by inductively coupled plasma spectroscopy (ICP, Table S1). Transmission electron microscopy (TEM) images of the catalysts were initially recorded. The V₂O₅ nanosheets exhibit an ultra-thin layered structure (Fig. 1(a)–(c)) [22]. The thickness of the V₂O₅ nanosheets was confirmed to be approximately six atomic layers (Fig. S1). Additionally, well-dispersed Pt NPs could be observed, which could help to fully expose Pt catalytic sites. The particle sizes of Pt NPs with different loading amounts are predominantly in the range of 4–5 nm (Fig. S2), illustrating that Pt NPs with specific particle sizes could be obtained using this synthesis procedure. High-resolution TEM (HR-TEM) images of Pt NPs-2/V₂O₅ were recorded (Figs. 1(d)–(f)). The observed *d* spacings of 0.215 and 0.227 nm correspond to the (102) and (111) facets of the V₂O₅

Fig. 2. (a) XPS spectrum of Pt NPs-2/V₂O₅ nanosheets; (b) V 2*p* core-level spectra of V₂O₅ nanosheets (A) and Pt NPs-2/V₂O₅ nanosheets (B); V 2*p* (c) and Pt 4*f* (d) core-level spectra of Pt NPs-2/V₂O₅ nanosheets.

Fig. 3. XRD patterns (a), and N_2 adsorption-desorption curves (b) of catalysts.

nanosheets and the (111) facets of face-centered cubic Pt NPs, respectively [23,24]. These results indicate that Pt NPs with particle sizes of approximately 4–5 nm could be uniformly dispersed on the surface of ultra-thin (with a thickness of approximately six atomic layers) 2D V_2O_5 nanosheets.

X-ray photoelectron spectroscopy (XPS) analyses of the catalysts were conducted. The detected elements, Pt, V and O (Fig. 2(a)), indicate the presence of both Pt NPs and V_2O_5 nanosheets in the catalyst. The V 2p high-resolution spectra of the V_2O_5 nanosheets and the Pt NPs-2/ V_2O_5 nanosheets are shown in Fig. 2(b). Here, two peaks attributed to V 2p_{3/2} and V 2p_{1/2} are observed. For the Pt NPs-2/ V_2O_5 nanosheets, the binding energies of the V 2p peaks shift to lower values compare with the V 2p peaks of the V_2O_5 nanosheets. This can be attributed to the partial reduction of V(V) to V(IV) due to the SMSIs between the V_2O_5 nanosheets and the Pt NPs [2,25]. Additionally, each peak in the XPS spectrum of the Pt NPs-2/ V_2O_5 nanosheets (shown in Fig. 2(b)) could be fitted into two peaks (Fig. 2(c)), and the peaks at approximately 516.3 and 523.6 eV correspond to V(IV) 2p_{3/2} and V(IV) 2p_{1/2}, respectively [11]. The detection of V(IV) can be ascribed to the transfer of electrons from the loaded Pt NPs to the V_2O_5 nanosheets. For the high-resolution spectrum of Pt 4f (Fig. 2(d)), each peak could be fitted into two peaks, corresponding to elemental Pt with zero (at approximately 71.0 and 74.2 eV) and two (at approximately 72.2 and 75.4 eV) valence electrons, respectively [26,27]. The generation of Pt(II) can also be ascribed to the transfer of electrons from the loaded Pt NPs to the V_2O_5 nanosheets. This phenomenon has been observed in many other catalyst systems consisting of PGMs and TMOs due to the formation of SMSIs [1–4].

To gain more information about the synthesized catalysts, XRD patterns of the catalysts were measured; these are shown in Fig. 3(a). Compared with the XRD pattern of bulk V_2O_5 (Fig. S3, PDF# 41–1426), only the diffraction peak of the (001) facets could be clearly observed (at approximately 20.1°) in the XRD pattern of the V_2O_5 nanosheets. This indicates the successful generation of ultra-thin 2D V_2O_5 nanosheets [20]. The XRD pattern of the Pt NPs-2/ V_2O_5 nanosheets also contains the diffraction peak corresponding to the V_2O_5 nanosheets; additionally, two diffraction peaks at approximately 39.8° and 46.3° were also detected, corresponding to the (111) and the (200)

facets of face-centered cubic Pt NPs (PDF# 88–2343), respectively [28]. Additionally, Raman spectra of the Pt NPs-2/ V_2O_5 nanosheets and the V_2O_5 nanosheets were collected; these are shown in Fig. S4. Here, characteristic peaks (assigned as A_g, B_{1g}, B_{2g} and B_{3g}) are observed [29], which are consistent with the vibrations of bulk V_2O_5 , illustrating the stable structure of the V_2O_5 nanosheets during the synthesis process. The SSAs of the V_2O_5 nanosheets and the Pt NPs-2/ V_2O_5 nanosheets were estimated through the Brunauer-Emmett-Teller method (BET, Fig. 3(b)). Compared with the SSAs of bulk V_2O_5 (2.3 m²/g), the SSAs of the V_2O_5 nanosheets increase to 19.8 m²/g, 8.6 times larger than that of bulk V_2O_5 . The SSAs of the Pt NPs-2/ V_2O_5 nanosheets are slightly lower (17.6 m²/g) than the SSAs of the V_2O_5 nanosheets, indicating that the structure of the V_2O_5 nanosheets is retained after the loading of Pt NPs. All of the obtained data suggests that the Pt NPs are loaded onto the surface of the V_2O_5 nanosheets uniformly and effectively.

The catalytic ability of the synthesized catalysts was measured by oxidizing DBT and its derivatives, in order to provide a green and feasible desulfurization method for these unreactive sulfides. Initially, sulfur removal using different reaction systems was conducted (Table 1, Fig. S5). Although a reasonable level of sulfur removal could be achieved (54.1%) after reaction for 5 h at 110 °C reaction temperature with V_2O_5 nanosheets, this is far away from deep desulfurization. When the V_2O_5 nanosheets were loaded with Pt NPs, the catalytic removal of sulfide improved significantly. Deep desulfurization (99.1%, < 5 μg g⁻¹) could be achieved using Pt NPs-2/ V_2O_5 nanosheets, meeting the latest regulations on sulfur content. Although a higher degree of sulfur removal could be achieved using the Pt NPs-3/ V_2O_5 nanosheets, long reaction time of 5 h were required to achieve deep desulfurization (Fig. S5). When bulk V_2O_5 is selected as a carrier, the degree of sulfur removal de-

Table 1
Sulfur removal with different reaction systems for 5 h.

Catalyst	Sulfur removal (%)
V_2O_5 nanosheets	54.1
Pt NPs-1/ V_2O_5 nanosheets	79.5
Pt NPs-2/ V_2O_5 nanosheets	99.1
Pt NPs-3/ V_2O_5 nanosheets	99.2
Pt NPs-2/bulk V_2O_5	19.7

Fig. 4. Sulfur removal of oils with different S-concentrations (a) and different substrates (b).

creases sharply, which might be ascribed to the smaller SSAs of bulk V_2O_5 ; only a tiny amount of Pt NPs could be loaded onto its surface. From a cost-effectiveness perspective, the Pt NPs-2/ V_2O_5 nanosheets were selected for further investigation. Additionally, the reaction temperature used in this study is lower than the temperatures used in many previously reported works (Table S2), indicating the superior catalytic ability of the as-prepared catalysts.

To investigate the feasibility for practical applications, we investigated the degree of sulfur removal from oils with different initial S-concentrations, different substrates and alkene/aromatic additives. As shown in Fig. 4(a), the degree of sulfur removal decreases slightly when the initial S-concentration increases from 500 to 1000 $\mu\text{g g}^{-1}$. In the system used here, abundant oxidant (O_2 in the air) could be provided, which is superior to reaction systems using H_2O_2 or organic peroxides as oxidants. For oils with different substrates (Fig. 4(b)), remarkable desulfurization performance was achieved. The degree of sulfur removal decreases in the order of: DBT > 4-MDBT > 4,6-DMDBT, which corresponds to the $f^2(r)$ Fukui functions of the sulfur atoms in these species (DBT (0.095) > 4-MDBT (0.076) > 4,6-DMDBT (0.070)) [30]. The methyl group in 4-MDBT would present an obstacle for the approach of the S atom to the generated active species, leading to a lower reaction activity. Thus, a far lower reaction activity would be expected for 4,6-DMDBT, which contains two methyl groups [31]. As shown in Fig. S6, cyclohexene and paraxylene were selected as model compounds and added to oils to give an

additive concentration of 20 wt%. Here, the lower degree of sulfur removal may be attributed to the competitive oxidation reactions between cyclohexene/paraxylene and DBT; this indicates that this reaction system is more suitable for oils containing lower concentrations of alkenes or aromatics.

Additionally, the stability and reusability of the catalyst are important considerations to be investigated. Thus, the performance of the catalyst over multiple reaction cycles was measured (Fig. 5(a)). The as-prepared catalyst could be recycled for at least seven reaction cycles without any significant loss of catalytic ability, exhibiting a desirable catalytic performance. The content of Pt NPs in the spent catalyst is approximately 1.81 wt% according to the ICP analysis, indicating that the leaching of Pt NPs could be effectively prevented due to the SMSIs. The TEM image (Fig. S7) of the spent catalyst also indicates that the supported Pt NPs could be encapsulated effectively on the surface of V_2O_5 nanosheets. The spent catalyst was also characterized by Fourier-transform infrared spectroscopy (FT-IR) measurements. As shown in Fig. 5(b), in addition to the retained characteristic peaks corresponding to V_2O_5 , a new peak appears at approximately 1162 cm^{-1} , which corresponds to S=O vibrations of sulfone that has precipitated in the catalyst phase. Additionally, the generation of sulfones was confirmed by gas chromatography-mass spectrometry analysis (GC-MS, Fig. S8). All of the observed data indicate that the as-prepared catalysts exhibit remarkable stability and reusability.

In summary, Pt NPs with well-defined particle sizes (approximately 4–5 nm) were loaded onto the surface of atom-

Fig. 5. (a) recycling performance of the as-prepared catalyst; (b) FT-IR analysis of fresh and spent catalyst.

ic-layered V_2O_5 nanosheets (approximately six atomic layers in thickness) effectively and uniformly. The obtained catalyst could successfully activate O_2 in the air under conditions of atmospheric pressure and 110 °C, providing a green and feasible desulfurization method. Unreactive sulfides in oil substrates could be successfully oxidized to polar sulfones, facilitating natural separation from the non-polar oils. It is noteworthy that the obtained catalyst exhibits desirable stability and reusability properties. This work presents a rational strategy to construct stable supported Pt NPs catalysts with remarkable catalytic ability, stability and reusability for many other aerobic oxidations.

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Graphical Abstract

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Pt nanoparticles encapsulated on V_2O_5 nanosheets carriers as efficient catalysts for promoted aerobic oxidative desulfurization performance

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Pt NPs with particle sizes of approximately 4–5 nm can be encapsulated effectively and uniformly on the surface of V_2O_5 nanosheets (with thickness of approximately six atomic layers) as promising catalysts for promoted aerobic oxidative desulfurization performance.

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V₂O₅纳米片固载Pt纳米粒子催化剂的构建及其增强催化空气氧化脱硫性能

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摘要: 以铂系金属为代表的贵金属催化剂在工业反应中通常表现出优异的催化性能,这是因为其具有独特的 d 带电子结构和较高的价电子比。近年来,由于大气排放法规愈发严苛,铂系贵金属催化剂在催化空气氧化燃油脱硫方面的研究引起了广泛关注。在该催化反应中,铂系金属纳米粒子可以有效活化空气中的氧气,产生的活性氧物种可以将油品中的噻吩类硫化物氧化为其对应的强极性砜类物质,从而可以将其从非极性的油品中分离出来,有效实现油品中硫化物的深度氧化脱除。然而,在反应过程中铂系贵金属纳米粒子易发生流失和烧结,从而导致催化剂的失活。因此,急需寻找一类可以有效固载铂系贵金属纳米粒子的载体。在目前已报道的众多载体中,以ZrO₂、TiO₂、CeO₂、ZnO等为代表的过渡金属氧化物引起了广泛的关注。通常认为,铂系贵金属纳米粒子的 d 轨道电子和过渡金属氧化物之间可形成金属-载体强相互作用。然而,目前所使用的过渡金属氧化物载体的比表面积较小,从而导致铂系贵金属纳米粒子难以有效且均匀地分散于其表面。

本文采用热膨胀气相剥离法制备了超薄V₂O₅纳米片,并通过超声辅助沉积法将Pt纳米粒子固载于其表面,从而得到一系列可高效活化空气氧化脱硫的催化剂(Pt NPs- n /V₂O₅纳米片)。通过电感耦合等离子体光谱、高倍透射电镜、原子力显微镜、X射线光电子能谱、X射线衍射、拉曼光谱和氮气吸附脱附等方法对催化剂的结构和形貌进行了表征。结果表明,尺寸为4–5 nm的Pt纳米粒子可有效均匀分散于层数约为6层的V₂O₅纳米片表面;在空气氧化脱硫反应中,当催化剂中Pt理论负载量为2 wt%时,反应5 h后,油品的脱硫率可达99.1%,实现了硫化物的深度氧化脱除。该反应体系对不同硫浓度、不同含硫底物的油品均有较好的脱除效果,但对含有烯烃、芳烃的油品脱除效果较差。此外,催化剂循环使用7次后,其脱硫活性仍无明显下降,表现出优异的重复使用性能。对反应后的催化剂进行表征,发现Pt几乎不发生流失,这可能是由于Pt纳米粒子和V₂O₅纳米片之间形成了金属-载体强相互作用。该结果为其他空气氧化反应的有效进行提供了新思路。

关键词: 氧化脱硫; 空气氧化; V₂O₅纳米片; Pt纳米粒子; 二苯并噻吩

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