




Editorial

Exclusive Papers on Environmentally Friendly Catalysis in China

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With the development of modern industries, environmental pollution has become a serious and urgent issue. Emissions of pollutants, such as volatile organic compounds (VOCs), methane, carbon monoxide, and nitrogen oxides (NO_x) as well as organics in wastewater, can cause environmental pollution to the atmosphere and water. Therefore, it is essential to develop effective technologies that can convert the above pollutants into harmless compounds. Among all of the methods developed to date, the catalytic approach is one of the pathways with the highest efficiency for the removal of pollutants.

This Special Issue on “Exclusive Papers on Environmentally Friendly Catalysis in China” includes 12 articles and 2 reviews, which show the novel development of catalysts, catalytic performance, and catalytic mechanisms for the removal of CH₄, CO, typical VOCs, NO_x, and organics in wastewater as well as the generation of hydrogen. In addition, although the works on the synthesis of unsaturated ether via direct and selective hydrogenation coupling of unsaturated aldehyde with alcohol and the synthesis of hex-anitrohexaazaisowurtzitane (HNIW) through hydrogenolytic debenzoylation of hexa-benzylhexaazaisowurtzitane (HBIW) and tetraacetyldibenzylhexaazaisowurtzitane (TADB) are not related to the topic of the removal of pollutants or the production of hydrogen, they still belong to the area of environmentally friendly catalysis. Hence, the above two articles are also included in this Special Issue.

Single-atom catalysis is a hot topic that has emerged in recent years, in which there is a maximization utilization efficiency of the active metal (especially precious metals) in a single-atom catalyst (SAC) [1]. Under practical conditions, however, the active sites and their stability against migration and aggregation are the critical issues. Yu et al. [2] reviewed the preparation and applications of single-atom catalysts in environmental catalysis, in which the preparation methods of SACs and their catalytic applications for the combustion of CH₄, CO, and VOCs and the reduction of NO_x were summarized. Moreover, future research trends and practical applications of SACs were also proposed. It was demonstrated that SACs have the advantages of more efficiency, more economic viability, and more uniqueness than their nanocluster or nanoparticle counterparts. Hence, reasonable designing of SACs with well-defined structures, high efficiency, good economic viability, and simple and controllable preparation strategies is of significance in future industrial applications.

Toluene is one of the typical VOCs, and its catalytic removal over CeO₂ with oxygen vacancies was investigated by Ismail et al. [3]. It was found that the CeO₂ catalyst with a shuttle-like morphology possessed a greater amount of oxygen vacancies and a higher



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surface area than those with the nanorod- and nanoparticle-like morphologies, thus allowing the former to exhibit better catalytic performance for toluene oxidation than the latter. Furthermore, the CeO₂ catalyst with a shuttle-like morphology showed good recyclability, long-term activity durability, and good water tolerance. Xue et al. [4] used quinary layered double hydroxide (CuCoMnNiFe-LDH)-derived single high-entropy spinel-type oxides (HEOs) as catalysts to investigate their catalytic behaviors in the oxidation of toluene, and found that the best activity and stability of the optimized HEO-600 catalyst derived after calcination at 600 °C were associated with the vast quantity of surface oxygen vacancies and the relative variable metal valence. The authors believed that they had developed a simple method to generate HEO materials effective for the application of VOC removal. Catalytic activity and water and sulfur dioxide tolerance are important for a catalyst used for the oxidative removal of VOCs. Sun et al. [5] prepared 0.46PdPt_{2.10}/V₂O₅-TiO₂, 0.41PdPt_{0.85}/V₂O₅-TiO₂, and 0.49PdPt_{0.44}/V₂O₅-TiO₂ catalysts using the hydrothermal and polyvinyl-alcohol-protecting NaBH₄ reduction approaches, and studied their catalytic activities for toluene oxidation. The characterization results revealed that the good catalytic performance of 0.46PdPt_{2.10}/V₂O₅-TiO₂ was related to its well-dispersed PdPt_{2.10} nanoparticles, higher amount of adsorbed oxygen species, good reducibility, large toluene adsorption capacity, and strong interaction between noble metals and support. The authors confirmed that vanadium was the main site for SO₂ adsorption while PdO was the secondary site for SO₂ adsorption, i.e., the doping of vanadium protected the active Pt site from being poisoned by SO₂, hence giving the 0.46PdPt_{2.10}/V₂O₅-TiO₂ catalyst good sulfur dioxide tolerance. An example of improving the sulfur dioxide resistance of catalysts is from the work carried out by Zhang et al. [6], who prepared reduced graphene oxide (rGO)-promoted α -MnO₂ nanorod-supported Pt (x Pt- y rGO/ α -MnO₂, x = 0.93 wt%, y = 0.5–2.0 wt%) catalysts using the polyvinyl-alcohol-protected reduction method. It was found that the strong metal-support interaction between Pt and α -MnO₂ favored the enhancement in catalytic activity for benzene oxidation; the doping of rGO to α -MnO₂ accelerated the charge migration and gave rise to a greater amount of benzene adsorption sites, thus improving the catalytic activity. In addition, the characterization results revealed that phenol, benzoquinone, and carboxylate species were the intermediates and were eventually converted into CO₂ and H₂O. In the presence of sulfur dioxide in the feedstock, the catalytic activity of α -MnO₂ decreased owing to the formation of manganese sulfate that covered part of the active sites, while loading Pt and rGO could hinder SO₂ chemisorption and prevent the active sites of the 0.94Pt-1.0rGO/ α -MnO₂ catalyst from being poisoned by SO₂, hence improving the SO₂ resistance of the catalyst. Ethyl acetate is one of the typical oxygen-containing VOCs. Bao et al. [7] fabricated y AuPd _{x} /TiO₂ (i.e., 0.35AuPd_{0.46}/TiO₂, 0.34AuPd_{2.09}/TiO₂, and 0.37AuPd_{2.72}/TiO₂) catalysts using the electrostatic adsorption approach and measured their catalytic activities for the oxidation of ethyl acetate. The authors assigned the high catalytic performance of 0.37AuPd_{2.72}/TiO₂ to the good dispersion of AuPd_{2.72} nanoparticles, good reducibility, large ethyl acetate adsorption capacity, and strong AuPd _{x} -TiO₂ interaction. The authors also detected the formation of main intermediates (e.g., acetaldehyde, ethanol, and acetic acid) in the oxidation of ethyl acetate and found that loading AuPd _{x} nanoparticles could effectively reduce the generation of the toxic acetaldehyde by-product. Moreover, the ethyl acetate oxidation mechanism over 0.34AuPd_{2.09}/TiO₂ was substantiated to follow the route of ethyl acetate → ethanol → acetic acid → acetate → carbon dioxide and water.

Methane is thought to be a non-VOC organic compound. In the past 100 years, the global warming potential of CH₄ has been reported to be 28–36 times higher than that of CO₂ [8–10]. Hence, it is imperative to control the emission of methane into the atmosphere. Zhang et al. [11] prepared Pd catalysts supported on Y-ZrO₂ with flower-like, spherical, reticulated, and bulk-specific morphologies, and found that the relatively large Pd particles and Pd⁰ jointly promoted the catalytic oxidation of CH₄ since the morphology influenced the microstructure of Y-ZrO₂ and the chemical state of the active Pd species.

The organics in wastewater can pollute the water environment. There are a number of methods to remove organics from wastewater, among which the photocatalytic approach is one of the most effective pathways. Jin et al. [12] constructed ZnS-based heterostructure photocatalysts (ZnO/ZnS) with ZnO and used them to degrade methyl orange, rhodamine B, and tetracycline. It was found that the high efficiency of the ZnO/ZnS photocatalyst for the degradation of methyl orange, rhodamine B, and tetracycline under UV light illumination was due to the improvement in light absorption ability, the effective separation of e^- and h^+ , and the synergistic interaction of ZnO and ZnS. Combined with the experimental and first-principles calculation results, the authors proposed a type II photocatalytic mechanism.

Since lean-burn combustion tends to emit excessive NO_x that is harmful to the atmospheric environment and human health, it is highly desirable to eliminate NO_x from automotive exhausts. Due to the high catalytic activity and N_2 selectivity as well as good hydrothermal stability, Cu^{2+} -exchanged zeolite (Cu-CHA) catalysts with a chabazite architecture are one of the most promising candidates for utilization as catalysts for the selective catalytic reduction of NO_x with ammonia (NH_3 -SCR). Zhao and coworkers [13] reviewed the active sites and the nature of the hydrothermal aging resistance and NH_3 -SCR reaction mechanisms over Cu-CHA catalysts, and, on this basis, the impacts of sulfate species, sulfate loading, emitted gaseous composition, and exposure temperature or time on the catalytic activity of Cu-CHA were discussed. The authors provided valuable insights into the matching between the design of NH_3 -SCR activity and sulfur dioxide resistance, and envisioned the challenges of NO_x removal over Cu-exchanged zeolites. In addition, sulfur poisoning diagnosis, effective regeneration approaches, and efficient catalyst designing for the aftertreatment system were also proposed to minimize the deterioration of Cu-CHA in the future. Chang et al. [14] prepared Cu/Ce deposited on pitch-based activated carbon fibers (ACF@Cu/Ce) and N-doped carbon nanofibers grown on the surface of ACF@Cu/Ce (N-CNF/ACF@Cu/Ce) and studied the NH_3 -SCR activity of these materials. It was observed that N-CNF/ACF@Cu/Ce showed NO conversions of 72–81% in a wide temperature range of 295–495 °C, indicating that such a material meets the requirements for practical industrial applications.

Water splitting into hydrogen over photocatalysts is an important pathway for obtaining clean and renewable energy, in which the key issue is the availability of high-performance photocatalysts. One of the main factors influencing the photocatalytic activity of a catalyst is the effective separation of the photogenerated charges. It is well known that the photoinduced charges can be separated efficiently at a heterointerface. Therefore, suitably designing a heterointerface between TiO_2 and another semiconductor would be an effective strategy for enhancing the photocatalytic performance of a catalyst [15]. Li et al. [16] fabricated durian-like $TiO_2@CdS$ core-shell photocatalysts using the solvothermal method in an ethylenediamine solution and investigated their H_2 production behaviors. The authors claimed that the as-obtained durian-like $TiO_2@CdS$ core-shell material overcame the disadvantages of single TiO_2 and CdS compounds, giving rise to a higher H_2 yield than pure TiO_2 (for example, the nanostructured $TiO_2@CdS$ derived at 180 °C for 16 h generated 1.5 times more H_2 than TiO_2). In addition, this durian-like $TiO_2@CdS$ photocatalyst also exhibited better stability than pure CdS. Dry reforming of methane (DRM) can convert CH_4 and CO_2 (two greenhouse gases) into syngas (CO and H_2) that can be directly used as raw material for the Fischer–Tropsch synthesis [17]. Hence, the DRM reaction can alleviate the energy crisis and improve the quality of the atmosphere. Chen et al. [18] carried out a study on catalytic DRM reaction over Ni/NiO supported on Ce-, Zr-, and Al-modified Y_2O_3 for hydrogen production, in which the authors found that the modified catalysts possessed an enhanced nickel-support interaction (giving rise to a better Ni dispersion) and improved alkalinity (beneficial for activating CO_2 and enhancing the catalytic activity). Moreover, the modified catalysts contained a greater amount of surface active oxygen ($O_2^{\delta-}$ and

O_2^-) species that favored the elimination of the deposited carbon species. In addition, the characterization results substantiated that the doping of a secondary metal could increase the DRM activity of the catalyst via promoting conversion of the formate intermediate species.

Unsaturated ether (UE) is a kind of high-grade ether that can be used as a solvent, perfume, medical anesthetic, and intermediate for pharmaceutical synthesis. There is a possibility to synthesize UE via the hydrogenation coupling of unsaturated aldehyde (UA) with alcohol [19,20]. Chen and coworkers [21] performed a study on the green and efficient synthesis of unsaturated ether from direct and selective hydrogenation coupling of unsaturated aldehyde with alcohol over bi-functional (hydrogenation activation and acid catalysis) Al-Ni-P heterogeneous catalysts. The authors noted that the bi-functional Al-Ni-P catalysts were efficient in the direct synthesis of UE (yield = 97%) from the selective hydrogenation coupling carbonyl of cinnamaldehyde or citral with $\text{C}_1\text{--C}_5$ primary or secondary alcohol at 0.1 MPa H_2 pressure and 120 °C. It is believed that such a UE synthesis strategy possesses several integrated advantages (e.g., high efficiency, green manner, and convenient operation), which would provide a potential for feasibly harvesting high-grade UE in the related synthesis of fine chemicals. Hydrogenolysis for N-debenzylation with H_2 over Pd/C or $\text{Pd}(\text{OH})_2/\text{C}$ is the most commonly used method due to its high efficiency, good atom economy, and easy-to-recycle active component [22]. Since HBIW is an important raw material for the synthesis of HNIW (CL-20), the hydrogenolytic debenzoylation of HBIW and TADB becomes a critical step for the synthesis of CL-20 [23,24]. Hence, developing high-efficiency catalysts around room temperature for the catalytic transformation of HBIW is an urgent requirement. Qiu and coworkers [25] conducted the hydrogenolytic debenzoylation of HNIW and TADB over $\text{Pd}(\text{OH})_2/\text{C}$ catalysts. It was found that all the $\text{Pd}(\text{OH})_2/\text{C}$ catalysts showed high catalytic activities in the debenzoylation of HBIW, but the catalytic activities of these catalysts in the debenzoylation of TADB were quite different. The authors concluded that the relatively high activity of Pd/HTC in the debenzoylation of TADB was related to the high Pd dispersion and novel mesoporous structure, whereas the higher activity and better stability of Pd/HTC- $\text{N}_{1:1}$ in the same reaction were associated with its good Pd dispersion, high total oxidized Pd species amount, and high surface pyridinic N content as well as the eggshell distribution of the Pd species on the support.

In summary, the above 14 articles published in this Special Issue are related to the preparation of various catalysts, catalytic activities, and/or catalytic reaction mechanisms for the elimination of CH_4 , CO, toluene, benzene, ethyl acetate, NO_x , and organics in wastewater as well as the production of H_2 from photocatalytic water splitting or dry reforming of methane. Furthermore, unsaturated ether was synthesized from direct and selective hydrogenation coupling of unsaturated aldehyde with alcohol, and HNIW was obtained from the hydrogenolytic debenzoylation of HNIW derivatives. At present, many efforts are being made related to catalysis in the removal of environmental pollutants and the controlled synthesis of useful organic products or intermediates.

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