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NH$_3$-SCR denitration catalyst performance over vanadium–titanium with the addition of Ce and Sb

Chi Xu$^1$, Jian Liu$^{1,*}$, Zhen Zhao$^1$, Fei Yu$^2$, Kai Cheng$^1$, Yuechang Wei$^1$, Aijun Duan$^1$, Guiyuan Jiang$^1$

1. State Key Laboratory of Heavy Oil, China University of Petroleum, Beijing 102249, China. E-mail: fxxuchi@126.com
2. Department of Agricultural and Biological Engineering, Mississippi State University, 130 Creelman St, Mississippi State, MS 39762, United States

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ABSTRACT

Selective catalytic reduction technology using NH$_3$ as a reducing agent (NH$_3$-SCR) is an effective control method to remove nitrogen oxides. TiO$_2$-supported vanadium oxide catalysts with different levels of Ce and Sb modification were prepared by an impregnation method and were characterized by X-ray diffractometer (XRD), Brunauer–Emmett–Teller (BET), Transmission electron microscopy (TEM), Fourier transform infrared spectroscopy (FT-IR), UV–Vis diffuse reflectance spectroscopy (UV–Vis DRS), Raman and Hydrogen temperature-programmed reduction (H$_2$-TPR). The catalytic activities of V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalysts for denitration were investigated in a fixed bed flow microreactor. The results showed that cerium, vanadium and antimony oxide as the active components were well dispersed on TiO$_2$, and the catalysts exhibited a large number of d–d electronic transitions, which were helpful to strengthen SCR reactivity. The V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalysts exhibited a good low temperature NH$_3$-SCR catalytic activity. In the temperature range of 210 to 400°C, the V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalysts gave NO conversion rates above 90%. For the best V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalyst, at a reaction temperature of 210°C, the NO conversion rate had already reached 90%. The catalysts had different catalytic activity with different Ce loadings. With the increase of Ce loading, the NO conversion rate also increased.

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Introduction

Recently, with the rapid development of society and gradual improvement of people’s living standards, environmental pollution as a byproduct of social progress has become more and more serious, especially the atmospheric pollution caused by human activities. Nitrogen oxides are one of the main hazardous atmospheric pollutants, and can cause environmental harm, such as the ozone depletion, acid rain and photochemical smog (Wang and Hao, 2012). Currently, among NO$_x$ emission control technologies, the technology of selective catalytic reduction (SCR) with NH$_3$ as the reducing agent is an effective way to remove NO$_x$ in the flue gas from fixed and mobile sources (Li et al., 2010). The SCR catalyst system is the core of NH$_3$-SCR technology. Using different SCR catalysts, the denitration activities show varying results.

At present, compared with other catalyst systems, the vanadium–titanium catalyst system has the advantages of being a mature production technology, with good anti-water and anti-sulfur resistance and high NO$_x$ removal efficiency (Li et al., 2011). Therefore, large commercial SCR catalysts are based on anatase TiO$_2$ carrier supported V$_2$O$_5$, modified by doping with WO$_3$ and MoO$_3$. Several different types are in use, including V$_2$O$_5$–WO$_3$/TiO$_2$, V$_2$O$_5$–MoO$_3$/TiO$_2$, V$_2$O$_5$–WO$_3$–MoO$_3$/TiO$_2$ and others.
(Kompio et al., 2012). However, the temperature for initiation of activity is relatively high for the vanadium and titanium catalyst system. Therefore, the SCR reactor needs to be installed before the dust collector, which can lead to catalyst deactivation caused by contaminated dust and other harmful impurities. In recent years, due to the potential advantages of low power consumption and low operating costs, low temperature SCR technology has attracted more and more attention from researchers. The development of a low-temperature, high denitrification activity vanadium–titanium based catalyst would have great significance.

Research has shown that CeO$_2$ loading on V$_2$O$_5$/TiO$_2$ catalysts can boost NH$_3$-SCR activity at low temperatures (Liu et al., 2012). First, CeO$_2$ has good redox properties (Ce$^{3+}$ ↔ Ce$^{4+}$). Secondly, CeO$_2$ has excellent oxygen storage capacity, abundant oxygen vacancies and oxygen ion transport characteristics, which can increase the oxygen adsorption and is conducive to the conversion of NO to NO$_2$ (Peng et al., 2012). Although much attention has been paid to the investigation of vanadium-based NH$_3$-SCR catalysts, few reports have appeared about the effect of different loading amounts of cerium and antimony on the TiO$_2$ support for NO reduction over a series of the V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalysts.

In this article, a series of vanadium–titanium based catalysts were modified by loading with cerium and antimony. The effects of metal ion doping on surface structure and morphology of catalysts as well as catalytic activity were investigated for different cerium and antimony loading amounts.

1. Experimental

1.1. Catalyst preparation

Titanium sulfate was dissolved in water. After vigorous stirring, aqueous ammonia was added dropwise to the solution until pH = 10. The solution was left to settle for 10 hr at room temperature and then was filtered. The precipitate was dried at 100°C for 10 hr, and then calcined at 550°C for 3 hr in static air to obtain the TiO$_2$ support. A series of V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalysts with fixed vanadium content of 5 wt.% and variable cerium and antimony loading amounts were prepared by the incipient-wetness impregnation method. After heating to dissolve ammonium metavanadate, cerium nitrate and antimony acetate were added to the solution, which was then stirred sufficiently at room temperature to form a mixed solution with given stoichiometry according to each molecular formula. Then the required amount of the above mixed solution was slowly added to TiO$_2$ support with thorough stirring at room temperature. The precursor was dried at 110°C for 12 hr, and then was heated to 100°C for 10 hr, and then calcined at 550°C for 3 hr in static air to obtain the TiO$_2$ support. The precursor was dried at 100°C for 10 hr, and then calcined at 550°C for 3 hr in static air to obtain the TiO$_2$ support.

1.2. Catalyst characterization

The crystal structures of the fresh catalysts were characterized by a powder X-ray diffractometer (Shimadzu XRD 6000, Japan) using Cu Ka ($\lambda = 0.154184$ nm) radiation with a nickel filter operating at 40 kV and 10 mA in the 2θ range of 10–80° at a scanning rate of 4°/min. The patterns were identified by comparison with reference patterns in the JCPDS database.

K$_2$ adsorption–desorption isotherms were measured at 77.35 K by a Micromeritics TriStar II 3020 analyzer (Micromeritics Instrument Corporation, USA). Prior to K$_2$ adsorption, all samples were degassed at 90°C for 1 hr and at 300°C for 8 hr under vacuum. The surface area was calculated by applying the Brunauer–Emmett–Teller (BET) method. The total pore volume (at a relative pressure of approximately 0.995), pore size distributions and average pore diameters were determined from the desorbed N$_2$ volume using the Barret–Joyner–Halenda (BJH) method. The transmission electron microscopy (TEM) images were observed using a JEOL JEM 2100 electron microscope equipped with a field emission source with an accelerating voltage of 200 kV. The powder samples were set in a transparent quartz cell and were measured in the region of 200 to 800 nm at room temperature, and pure BaSO$_4$ was used as reference. Fourier Transform infrared (FT-IR) absorbance spectra were recorded in the wavenumber range from 6000 to 400 cm$^{-1}$ via a FTS-3000 spectrophotometer (Digilab Company, USA). The resolution was set at 2 cm$^{-1}$. A pellet was produced by mixing the sample with KBr (1:100 weight ratio). The UV–Vis diffuse reflectance spectroscopy (UV–Vis DRG) experiments were carried out on a UV–Vis spectrophotometer (U-4100, Hitachi Ltd., Japan) with an integrating sphere diffuse reflectance attachment. UV-Raman experiments were performed at room temperature with a Renishaw Micro-Raman System 2000 spectrometer (Renishaw Company Ltd., UK). A laser radiating at 532 nm was used as the excitation source. Spectral resolution was 2 cm$^{-1}$. Hydrogen temperature-programmed reduction (H$_2$-TPR) analyses were carried out on a Quantachrome apparatus (Autosorb-iQ, USA).

1.3. Catalytic activity measurement

The NH$_3$-SCR activities of a series of V$_5$Ce$_x$Sb$_y$/TiO$_2$ catalysts were measured in a fixed bed quartz micro-reactor in steady flow mode. The required mass of catalysts was 0.4 g at 40–60 mesh size for each test. The reactant gas mixture consisted of 1000 ppm NO, 1000 ppm NH$_3$, 3% O$_2$ and balance N$_2$. The total flow rate was 500 mL/min. The GHSV was 45,000 h$^{-1}$. The hydrogen consumption signal was monitored by a TCD. Before the outlet gases entered the TCD, a cooling trap and a filter packed with molecular sieve 5 A (60–80 mesh) were used to remove H$_2$O and CO$_2$.

\[
\text{NO conversion} = \left(\frac{[\text{NO}]_{\text{inlet}} - [\text{NO}]_{\text{outlet}}}{[\text{NO}]_{\text{inlet}}}\right) \times 100\%
\]

where $[\text{NO}]_{\text{inlet}}$ represents the NO concentration in the inlet (ppm), and $[\text{NO}]_{\text{outlet}}$ represents the NO concentration in the outlet (ppm).
The $N_2$ selectivity in the SCR reaction was calculated from Eq. (2):

$$N_2\text{selectivity} = \left(1 - \frac{2[N_2O]_{\text{outlet}} + [NO]_{\text{inlet}} - [NO]_{\text{outlet}} - [NH_3]_{\text{outlet}}}{[NO]_{\text{inlet}} + [NH_3]_{\text{inlet}} - [NO]_{\text{outlet}} - [NH_3]_{\text{outlet}}} \right) \times 100\%.$$  

### 2. Results and discussion

#### 2.1. Results of XRD

XRD patterns of $V_5Ce_xSb_y/TiO_2$ catalysts with different Ce and Sb loadings are shown in Fig. 1. All the samples exhibit similar characteristics for the main diffraction peaks. A slight difference can be observed in peak intensities. Diffraction peaks at $2\theta = 25, 38$ and $48^\circ$ can be observed, which are attributed to the characteristic peaks of anatase TiO$_2$. Anatase TiO$_2$ has good activity for NH$_3$-SCR (Alemany et al., 1996). Moreover, no peaks for vanadium or antimony oxides species are observed in XRD patterns of the $V_5Ce_xSb_y/TiO_2$ catalysts. These results show that the vanadium and antimony oxide species are well dispersed on the surface of TiO$_2$ support, and no crystal phase V$_2$O$_5$ was observed, which may be helpful to enhance NH$_3$-SCR activity. Weak diffraction peaks can be observed at $2\theta = 28$ and $33^\circ$, which are attributed to the characteristic peaks of CeO$_2$ (Fig. 1).

#### 2.2. Results of BET characterization

The BET specific surface area, the BJH desorption pore volume and the average pore diameter of pure TiO$_2$ and $V_5Ce_xSb_y/TiO_2$ catalysts are shown in Table 1. As can be observed from this table, the series of $V_5Ce_xSb_y/TiO_2$ catalysts have large specific surface areas, which may enhance the NH$_3$-SCR reaction catalytic activity (Liu et al., 2008, 2009). Compared with the pure TiO$_2$ support, the specific surface area and pore volume decrease with cerium and antimony incorporation. These trends are attributed to the coverage of the external surface and blocking of the pores of the TiO$_2$ support after loading the active components, respectively. However, with increasing Ce loading and addition of antimony, the specific surface area of $V_5Ce_xSb_y/TiO_2$ catalysts increased. This may be presumably due to the strong interaction between vanadium, titanium, cerium and antimony species, which is beneficial for the good dispersion of active components on the titania support (Liu et al., 2008; Liu and He, 2010).

### Table 1 – Physical and morphological properties of pure TiO$_2$ and $V_5Ce_xSb_y/TiO_2$ catalysts.

<table>
<thead>
<tr>
<th>Samples</th>
<th>BET surface area (m$^2$/g)</th>
<th>Pore volume (cm$^3$/g)</th>
<th>Average pore diameter (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO$_2$</td>
<td>60.4</td>
<td>0.2591</td>
<td>13.436</td>
</tr>
<tr>
<td>$V_5Ce_{15}/TiO_2$</td>
<td>44.6</td>
<td>0.2013</td>
<td>12.603</td>
</tr>
<tr>
<td>$V_5Ce_{25}/TiO_2$</td>
<td>46.0</td>
<td>0.1867</td>
<td>11.261</td>
</tr>
<tr>
<td>$V_5Ce_{35}/TiO_2$</td>
<td>50.3</td>
<td>0.1999</td>
<td>11.743</td>
</tr>
<tr>
<td>$V_5Ce_{35Sb_2}/TiO_2$</td>
<td>50.5</td>
<td>0.1905</td>
<td>11.262</td>
</tr>
</tbody>
</table>

### Fig. 2 – $N_2$ adsorption–desorption isotherms (a) and the Barret–Joyner–Halenda (BJH) desorption pore size distribution (b) of pure TiO$_2$ and $V_5Ce_xSb_y/TiO_2$ catalysts with different Ce and Sb loadings.
Fig. 2a shows the N$_2$ adsorption–desorption isotherms of pure TiO$_2$ and V$_x$Ce$_y$Sb/TiO$_2$ catalysts tested at liquid nitrogen temperature. According to the BDDT classification, the adsorption isotherms of the samples are similar to type IV isotherms. These types are associated with capillary condensation in the mesoporous range (2–50 nm). The hysteresis loops belong to typical type E, indicating the existence of an “ink-bottle-shaped” pore structure (Liu et al., 2010; Liu and He, 2010). Fig. 2b shows the BJH desorption pore size distributions, indicating that the active crystallite phases in the prepared catalysts mainly have mesopores with distribution peaks near 12 nm. It can be seen that the V$_x$Ce$_y$/TiO$_2$, V$_x$Ce$_y$/TiO$_2$ and V$_x$Ce$_y$/Sb$_2$/TiO$_2$ catalysts have more abundant and smaller mesopores than V$_x$Ce$_y$/TiO$_2$, which can supply more active sites and inner surface area for the NH$_3$-SCR reaction.

2.3. TEM images of catalysts

Fig. 3 presents TEM images of V$_x$Ce$_y$Sb$_2$/TiO$_2$ catalysts with different Ce and Sb loadings. The particle size and morphologies of the catalyst samples can be observed from the TEM images. No obvious pore agglomeration can be seen. The TEM image of V$_x$Ce$_y$/TiO$_2$ illustrates that the typical particles are well dispersed with sizes in the range 20–50 nm. V$_x$Ce$_y$/TiO$_2$ shows crystalline sizes of 15–40 nm, and V$_x$Ce$_y$/TiO$_2$ presents slightly smaller crystalline sizes of 10–35 nm. Furthermore,
V₅Ce₃₅Sb₂/TiO₂ shows the smallest particle sizes at around 5–30 nm, and good dispersion.

### 2.4. Results of FT-IR characterization

FT-IR spectra of V₅CeₓSbᵧ/TiO₂ catalysts with different Ce and Sb loadings are shown in Fig. 4. Compared with the TiO₂ support, the spectra of V₅CeₓSbᵧ/TiO₂ catalysts show similar structure characteristics, and the band at 500–540 cm⁻¹ is assigned to the anatase TiO₂, suggesting that the skeleton structure of TiO₂ does not change dramatically after loading the metal oxides. These results show that the interaction of the TiO₂ support and metal species is relatively stable after the V₅CeₓSbᵧ/TiO₂ catalysts are calcined at 550°C. Characteristic vibration peaks do not appear for the active components, which is due to the cerium, vanadium and antimony oxide infrared absorption peaks being obscured by the strong signal of the TiO₂ support.

### 2.5. Results of UV–Vis DRS characterization

Fig. 5 shows UV–Vis DRS spectra of V₅CeₓSbᵧ/TiO₂ catalysts with different Ce and Sb loadings. The band at about 230 nm is assigned to the bulk vanadia and is formed by charge transfer between the valence band of O₂⁺ and the empty orbital V₅d (Larrubia et al., 2000). In contrast to that of the TiO₂ support, the intense band at 350 nm has an obvious red shift, which is due to the charge transfer between the conduction band or the valence band of active metal and TiO₂ (Nagaveni et al., 2004). Therefore, the band gap of TiO₂ is reduced, leading to a red shift in the absorption edge of TiO₂. The band for V₅CeₓSbᵧ/TiO₂ catalysts centered at 550 nm is assigned to cerium oxide. For antimony doping, the band (e line in Fig. 5) at 550 nm disappears, which is caused by substitution of antimony oxide in the cerium oxide position. A broad band between 500 and 800 nm is assigned to d-d transitions for V₅CeₓSbᵧ/TiO₂ catalysts, which is helpful to enhance the redox properties of the catalysts and improve the activity of the SCR reaction.

### 2.6. Results of Raman spectroscopy

The Raman spectra of pure TiO₂ and V₅CeₓSbᵧ/TiO₂ catalysts with the excitation line at 532 nm are shown in Fig. 6. The bands at 146, 396, 515 and 648 cm⁻¹ are assigned to typical Raman peaks of the TiO₂ support (Fig 6a). The intense band at 146 cm⁻¹ is assigned to the bending vibration mode of O–Ti–O. The weak bands at 396 and 515 cm⁻¹ are assigned to the symmetric and unsymmetric bending vibration of O–Ti–O, respectively. The weak band at 648 cm⁻¹ is attributed to the O–Ti–O symmetric stretching vibration. Because the Raman signals of the support are strong, the Raman shift of CeO₂ species cannot be well discriminated between 100 and 1200 cm⁻¹. Thus, the Raman spectra are carefully analyzed between 300 and 700 cm⁻¹ in Fig. 6b. For the V₅CeₓSbᵧ/TiO₂ catalyst, the vibration peak of Ce–O at 465 cm⁻¹ does not appear. However, starting from V₅Ce₂₅TiO₂, the band at 465 cm⁻¹ appears and the intensity of this peak increases with increasing Ce content even though the intensity of the band is weak. Moreover, the Raman peaks of V and Sb oxide species are not found in the Raman spectra, which is consistent with the results of XRD. The result also demonstrates that vanadium and antimony oxide species have low content and are dispersed well on the surface of the TiO₂ support.

### 2.7. Results of H₂-TPR

To gain a better understanding of the redox properties of the catalysts, the H₂-TPR of pure TiO₂ and V₅CeₓSbᵧ/TiO₂ catalysts were measured, as shown in Fig. 7. The maximum reduction peak of TiO₂ appeared at 560°C. All V₅CeₓSbᵧ/TiO₂ catalysts showed two major reduction peaks between 300 to 600°C and 660 to 730°C, which could be attributed to the reduction of surface vanadia, ceria and antimony species, respectively. These surface reduction peaks are assigned to the reductions from V⁵⁺ to V³⁺, Ce⁴⁺ to Ce³⁺ and Sb⁵⁺ to Sb³⁺ (Besselmann et al., 2001). With increasing Ce loading, a small shoulder reduction peak appears at 770°C for all V₅CeₓSbᵧ/TiO₂ catalysts, which is assigned to reduction of lattice oxygen (Lee et al., 2014). The reduction profiles of the series of V₅CeₓSbᵧ/TiO₂ catalysts strongly depend on the Ce and Sb loadings. As the Ce and Sb contents increase in the catalysts, the low temperature reduction peak shifts to higher temperature (from 470°C for V₅Ce₁₅/TiO₂ to 526°C for V₅Ce₃₅Sb₂/TiO₂). The second reduction peak also shifts toward higher temperature with increasing Ce content (from 689°C for V₅Ce₁₅/TiO₂ to
704°C for V5Ce35/TiO2). The temperatures of different peaks in the TPR profiles indicate the metal-support interaction over the supported catalysts. According to the above analyses, it can be seen that Ce and Sb may optimize the interaction between the support and active metals and improve the dispersion of active species, as confirmed by the above TEM characterization of the V5CeSb/TiO2 catalysts.

2.8. Activity measurements

A series of V5CeSb/TiO2 catalysts were tested in the NH3-SCR of NO and the results are shown in Fig. 8a. In these tests, all vanadium–titanium based catalysts had good low temperature NH3-SCR activity for different Ce and Sb loadings. At the reaction temperature of 225°C, NO conversion reached 90% over all the catalysts. The NO conversion increased with increasing Ce loading. This may be due to the fact that addition of Ce and Sb can promote the oxidation of vanadium at low valence, thus improving the ability of absorption, storage and transfer for surface oxygen (Cousin et al., 2007). At 225–400°C, NO conversion could be maintained above 90% for all V5CeSb/TiO2 catalysts with a wide temperature window. Among all the catalysts tested in this study, the V5CeSb/TiO2 catalyst exhibited the best catalytic activity, and NO conversion had already reached 90% at 210°C. The addition of antimony (2 wt.%) led to the improvement of the low temperature NH3-SCR activity. Therefore, at low temperature, the NH3-SCR activities of the catalysts follow the sequence V5Ce35/TiO2 < V5Sb25/TiO2 < V5Ce35Sb2/TiO2 < V5Ce35/TiO2 < V5–Ce35Sb2/TiO2. The results suggest that increasing cerium and addition of antimony can especially improve the catalytic activity of the SCR reaction at low temperature.

The selectivity of N2 over V5CeSb/TiO2 catalysts is shown in Fig. 8b. It can be seen that the formation of N2 was decreased above 300°C for all catalysts. With increasing Ce loading, the N2 selectivity slowly increased. This indicates that high contents of cerium oxide were mainly responsible for the enhanced NH3 activation. The addition of antimony (2 wt.%) resulted in the highest maximum N2 selectivity compared to V5Ce/TiO2 catalysts. It is worth noting that the V5CeSb/TiO2 catalyst exhibited high N2 selectivity (>90%) until the temperature was as high as 500°C. Among the four catalysts, V5Ce35Sb2/TiO2 gave the highest NO conversion and high N2 selectivity over a wide temperature range of 225–400°C. From Fig. 8a and b, it can be seen that both the SCR activity and N2 selectivity are mainly dependent on the Ce and Sb contents of V5CeSb/TiO2 catalysts.

3. Conclusions

A series of V5CeSb/TiO2 catalysts were prepared by the incipient-wetness impregnation method. According to the results of XRD, both vanadium and antimony oxide species on the surface of TiO2 support have good dispersion, which is important to improve NH3-SCR activity. The different Ce and Sb contents of vanadium–titanium based catalysts have different influences on NH3-SCR reactivity at low temperature. The best catalytic activity was obtained over the V5CeSb/TiO2 catalyst and its lowest activity temperature for 90% NO conversion was 210°C. With the increase of cerium content and the addition of antimony, the catalytic activity at

Fig. 7 – H2 TPR profiles of V5CeSb/TiO2 catalysts with different Ce and Sb loadings.

Fig. 8 – NO conversion (a) and N2 selectivity (b) as a function of reaction temperature over V5CeSb/TiO2 catalysts with different Ce and Sb loadings.
low temperature and N₂ selectivity at high temperature could be improved. This indicates that doping with Ce and Sb is a promising and efficient way to enhance the catalytic and selectivity activity of NH₃-SCR at low temperature.

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  National Chiao Tung University  
  Taiwan, China

- **Ng Wun Jern**  
  Nanyang Environment & Water Research Institute, Singapore

- **Clark C. K. Liu**  
  University of Hawaii at Manoa, USA

- **Zijian Wang**  
  Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, China

- **Zhiliang Xiao**  
  Shandong University, China

- **Min Yang**  
  Queen’s University, Canada

- **Daqiang Yin**  
  Tongji University, China

- **Zhongtang Yu**  
  The Ohio State University, USA

#### Environmental toxicology and health

- **Jingwen Chen**  
  Dalian University of Technology, China

- **Jianying Hu**  
  Peking University, China

- **Guibin Jiang**  
  Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, China

- **Tsuyoshi Nakashima**  
  Gifu Pharmaceutical University, Japan

- **Willie Peijnenburg**  
  University of Leiden, The Netherlands

- **Bingsheng Zhou**  
  Institute of Hydrobiology, Chinese Academy of Sciences, China

- **Hong He**  
  Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, China

- **Junhua Li**  
  Tsinghua University, China

- **Wenfeng Shangguan**  
  Shanghai Jiao Tong University, China

#### Environmental catalysis and materials

- **Christopher Anderson**  
  Massey University, New Zealand

- **Zacong Cai**  
  Nanjing Normal University, China

- **Xinbin Feng**  
  Institute of Geochemistry, Chinese Academy of Sciences, China

- **Hongjing Hu**  
  Hua Zhong Agricultural University, China

- **Kia-Che Lam**  
  The Chinese University of Hong Kong  
  Hong Kong, China

- **Erwin Klumpp**  
  Research Centre Juelich, Agrosphere Institute  
  Germany

- **Environmental biology**

- **Yong Cai**  
  Florida International University, USA

- **Henner Holloer**  
  RWTH Aachen University, Germany

- **Jae-Seong Lee**  
  Sungkyunkwan University, South Korea

- **Christopher Rensing**  
  University of Copenhagen, Denmark

- **Bojan Sedmak**  
  National Institute of Biology, Slovenia

- **Lirong Song**  
  Institute of Hydrobiology, Chinese Academy of Sciences, China

- **Chunxia Wang**  
  National Natural Science Foundation of China

- **Gehong Wei**  
  Northwest A & F University, China

#### Environmental analysis and method

- **Zongwei Cai**  
  Hong Kong Baptist University, Hong Kong, China

- **Zhiping Chen**  
  Dalian Institute of Chemical Physics, Chinese Academy of Sciences, China

- **Minghui Zhang**  
  Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, China

- **Environmental solid waste and green chemistry**

- **Pinjing He**  
  Tongji University, China

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