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## **1. Main Research Results**

### **Photocatalytic Reduction of CO<sub>2</sub> with Oxide Photocatalyst**

Converting of greenhouse gas CO<sub>2</sub> into useful hydrocarbon fuel by photocatalytic reaction is expected as an ultimate technology to solve the global warming problem. However, the conversion efficiency is still very low, because reduction of CO<sub>2</sub> needs a good adsorption property of the material to CO<sub>2</sub>, a very strong reduction potential, a multi-electron reaction process, etc. In this study, we have developed a simple ion exchange method for synthesis of micro/mesoporous Zn<sub>2</sub>GeO<sub>4</sub> with crystalline pore-walls at room temperature. Such prepared mesoporous Zn<sub>2</sub>GeO<sub>4</sub> showed 6 times higher photoactivity for converting CO<sub>2</sub> into CH<sub>4</sub> fuel in contrast to the sample prepared by solid state route, since large specific surface area of the mesoporous photocatalyst are beneficial to CO<sub>2</sub> capture and conversion. In addition, the evolution rate of CH<sub>4</sub> over micro/mesoporous Zn<sub>2</sub>GeO<sub>4</sub> could be significantly enhanced by loading Pt as co-catalyst, because the co-catalyst promotes the multi-electron reaction process which benefits the CO<sub>2</sub> photoreduction.

## References

- ① Ning Zhang, Shuxin Ouyang, Peng Li, Yuanjian Zhang, Guangcheng Xi, Tetsuya Kako, and Jinhua Ye, "Ion-Exchange Synthesis of Micro/Mesoporous Zn<sub>2</sub>GeO<sub>4</sub> Photocatalyst at Room Temperature for Photoreduction of CO<sub>2</sub>", *Chem. Commun.* 47, 2041-2043, 2011.
- ② S. C. Yan, S. X. Ouyang, J. Gao, M. Yang, J. Y. Feng, X. X. Fan, L. J. Wan, Z. S. Li, J. H. Ye, Y. Zhou and Z. G. Zou. "A Room-Temperature Reactive Template Route to Mesoporous ZnGa<sub>2</sub>O<sub>4</sub> with Improved Photocatalytic Activity in Reduction of CO<sub>2</sub>", *Angew. Chemie*, 49, 6400-6404, 2010.

## **2. List of Publications:**

- 1) Ning Zhang, Shuxin Ouyang, Peng Li, Yuanjian Zhang, Guangcheng Xi, Tetsuya Kako, and Jinhua Ye, "Ion-Exchange Synthesis of Micro/Mesoporous Zn<sub>2</sub>GeO<sub>4</sub> Photocatalyst at Room Temperature for Photoreduction of CO<sub>2</sub>", *Chem. Commun.* 47, 2041-2043, 2011.
- 2) S. C. Yan, S. X. Ouyang, J. Gao, M. Yang, J. Y. Feng, X. X. Fan, L. J. Wan, Z. S. Li, J. H. Ye, Y. Zhou and Z. G. Zou. "A Room-Temperature Reactive Template

- Route to Mesoporous  $\text{ZnGa}_2\text{O}_4$  with Improved Photocatalytic Activity in Reduction of  $\text{CO}_2$ ", *Angew. Chemie*, 49, 6400-6404, 2010.
- 3) H. Shi, Z. Li, J. Kou, J. Ye, Z. Zou, "Facile Synthesis of Single-Crystalline  $\text{Ag}_2\text{V}_4\text{O}_{11}$  Nanotube Material as a Novel Visible-Light-Sensitive Photocatalyst", *J. Phys. Chem. C*, 115(1), 145-151, 2011
  - 4) Xiaoqing Chen, Zhaosheng Li, Jinhua Ye, Zhigang Zou, A Forced Impregnation Approach to Fabrication of Large-area Three-dimensionally Ordered Macroporous Metal Oxides, *Chem. Mater* 22, 3583-3585 (2010).
  - 5) Shuxin Ouyang, Di Chen, Defa Wang, Zhaosheng Li, Jinhua Ye, Zhigang Zou, From  $\beta$ -phase particle to  $\alpha$ -phase hexagonal-platelet superstructure over  $\text{AgGaO}_2$ : phase transformation, formation mechanism of morphology, and photocatalytic properties, *Crystal Growth & Design* 10(7), 2921-2927 (2010).
  - 6) Jun Lv, Tetsuya Kako, Zhaosheng Li, Zhigang Zou, and Jinhua Ye, Synthesis and Photocatalytic Activities of  $\text{NaNbO}_3$  Rods Modified by  $\text{In}_2\text{O}_3$  Nanoparticles, *J. Phys. Chem* 114, 6157-6162 (2010).
  - 7) Jun Lv, Tetsuya Kako, Zhigang Zou and Jinhua Ye, Enhanced N-doping Efficiency and Photocatalytic  $\text{H}_2$  Evolution Rate of  $\text{InNbO}_4$  by Mechanochemical Activation, *J. Mater. Res* 25(1), 159-166 (2010).
  - 8) Qiuye Li, Tetsuya Kako, Jinhua Ye,  $\text{PbS/CdS}$  nanocrystals-sensitized titanate network films: enhanced photocatalytic activities and super-amphiphilicity, *J. Mater. Chem* 20(45), 10187-10192 (2010).
  - 9) Xiukai Li, Bing Yue, Jinhua Ye, Photocatalytic Hydrogen Evolution over  $\text{SiO}_2$ -Pillared and Nitrogen-Doped Titanic Acid under Visible Light Irradiation, *Applied Catalysis A: General* 390, 195-200 (2010).
  - 10) Yingpu Bi and Jinhua Ye, Direct Conversion of Commercial Silver Foils into High Aspect Ratio  $\text{AgBr}$  nanowires with Enhanced Photocatalytic Properties, *Chem. Eur. J* 16(34), 10327-10331 (2010).
  - 11) Yingpu Bi and Jinhua Ye, Cold-welding fabrication of highly ordered gold nanochannel monolayers in aqueous medium, *Chem. Commun* 46(37), 6912-6914 (2010).
  - 12) Guangcheng Xi and Jinhua Ye, "Synthesis of Hierarchical Macro-/Mesoporous Solid Solution Photocatalysts by a Polymerization-Carbonization-Oxidation route: The case of  $\text{Ce}_{0.49}\text{Zr}_{0.37}\text{Bi}_{0.14}\text{O}_{1.93}$ ", *Chem. Eur. J*, 16, 8719-8725, 2010.
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- 14) Guangcheng Xi and Jinhua Ye, "Synthesis of bismuth vanadate nanoplates with exposed {001} facets and enhanced visible-light photocatalytic properties", *Chem. Commun.*, **46**, 1893-1895, 2010.
- 15) Junyu Cao, Tetsuya Kako, Naoki Kikugawa, Jinhua Ye, Photoanodic properties of pulsed-laser-deposited  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> electrode, *J Phys. D: Appl. Phys* 43, 325101 (2010).
- 16) Hua Tong and Jinhua Ye, Building Niobate Nanoparticles with Hexaniobate Lindqvist Ions, *European Journal of Inorganic Chemistry* 1473-1480 (2010).
- 17) Qiuye Li, Tetsuya Kako, Jinhua Ye, Strong adsorption and effective photocatalytic activities of one-dimensional nano-structured silver titanates, *Applied Catalysis A: General* 375, 85-91 (2010).
- 18) Qiuye Li, Bing Yue, Hideo Iwai, Tetsuya Kako, Jinhua Ye Carbon Nitride Polymers Sensitized with N-doped Tantalum Acid for Visible Light-Induced Photocatalytic Hydrogen Evolution, *J. Phys. Chem. C* 114, 4100-4105 (2010).
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- 21) Z. G. Yi and Jinhua Ye, Iwai, Hideo Photochromism and visible light induced H<sub>2</sub> generation in Sr<sub>2</sub>TiO<sub>4</sub>:Cr Complexes, *Appl. Phys. Lett* 96, art. 114103 (2010).
- 22) Tetsuya Kako, Weifeng Yao, and Jinhua Ye, Preparation and characterization of visible light sensitive Fe- and Ta-codoped TiO<sub>2</sub> photocatalyst, *J. Mater. Res* 25(1), 110-116 (2010).
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- 25) Yuanjian Zhang, Toshiyuki Mori, Jinhua Ye, Markus Antonietti, "Phosphorus-Doped Carbon Nitride Solid: Enhanced Electrical Conductivity and Photocurrent Generation", *JACS*, **132(18)**, 6294-6295, 2010.

### **3. Conference Presentations (International & Domestic)**

#### Invited talks

- 1) Jinhua Ye, Nano Photocatalytic Materials: possibilities & challenges, Australia-China symposium on nanomaterials for clean energy, Queensland University, Brisbane, Australia, 2010/05/27 -29
- 2) Jinhua Ye, Nano Photocatalytic Materials: possibilities & challenges, 1st International Conference on Materials for Energy, DECHEMA Congress Office Karlsruhe Convention Center, Karlsruhe, Germany, 2010/07/04 – 08
- 3) Jinhua Ye, Zhiguo Yi, Tetsuya Kako, New Visible Light Active Photocatalyst Ag<sub>3</sub>PO<sub>4</sub>, 10<sup>th</sup> Meeting of Photo Functionalized Materials Society, Research Center for Advanced Science and Technology, Tokyo, Japan, 2010/07/14
- 4) Jinhua Ye, Zhiguo Yi, Tetsuya Kako, Naoki Kikugaw, Shuxin Ouyang, Naoto Umezawa, Zhigang Zou, Nano Photocatalysts for Solar Chemical Conversion and Environmental Remediation, IUMRS-ICA2010, International Union of Materials Research Societies, Qingdao International Convention Center, Qingdao, China, 2010/09/25 – 28
- 5) Naoto Umezawa, Shuxin Ouyang, Jinhua Ye, Theoretical study of an excellent photocatalyst Ag<sub>3</sub>PO<sub>4</sub>, IUMRS-ICA2010, International Union of Materials Research Societies, Qingdao International Convention Center, Qingdao, China, 2010/09/25 – 28
- 6) Kikugawa Naoki, Zhiguo Yi, Tetsuya Kako, Shuxin Ouyang, Junyu Cao, Naoto Umezawa, Jinhua Ye, Photo-induced Oxidization Properties of a Silver Orthophosphate under Visible-light Irradiation, 4th International Workshop on Advanced Ceramics, Nagoya Institute of Technology (Nagoya, Aichi), 2010/12/10-12

#### Oral Presentation

- 7) Naoto Umezawa, Jinhua Ye, EFFECTS OF LOADING MULTIVALENT OXIDES ON PHOTOCATALYSTS, 18th International Conference on Photochemical Conversion and Storage of Solar Energy (IPS-18), Korea University, Seoul, Korea, 2010/07/25 - 30
- 8) Zhiguo Yi, Shuxin Ouyang, Naoto Kikugawa, Tetsuya kako, Jinhua Ye, Highly Effective O<sub>2</sub> Evolution over a Novel Photocatalyst Ag<sub>3</sub>PO<sub>4</sub> under Visible-light Irradiation, National Conference on Solar Energy Photochemistry and Photocatalysis, Photochemical Professional Committee of Chinese Renewable Energy, University of Yunnan, Kunming, China, 2010/08/05-06

- 9) Yingpu Bi, Jinhua Ye, Facile synthesis of one-dimensional metal/semiconductor hetero-nanostructures and their catalytic properties, National Conference on Solar Energy Photochemistry and Photocatalysis, Photochemical Professional Committee of Chinese Renewable Energy, University of Yunnan, Kunming, China, 2010/08/05-06
- 10) Tetsuya Kako, Jinhua Ye, Degradation of ethanal by ilmenite- and pyrochlore-structured silver antimonate photocatalysis, Pachifichem 2010, Convention center in Hawaii, Honolulu, USA, 2010/12/15-20
- 11) ZHANG Yuanjian, Toshiyuki Mori, Jinhua Ye, Markus Antonietti, Photovoltaic system based on polymeric carbon nitride solids, Pachifichem 2010, Convention center in Hawaii, Honolulu, USA, 2010/12/15-20
- 12) Tetsuya Kako, Qiuye LI, and Jinhua Ye, Photophysical and Photocatalytic Properties of Ti-based Oxides, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabandai, Fukushima, 2011/1/20-22
- 13) Hua Tong, Naoto Umezawa and Jinhua Ye, Electronic Coupling Assembly of Titanium Oxide Nanocrystals: Self-Narrowed Band Gap and Photocatalytic Application, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabandai, Fukushima, 2011/1/20-22
- 14) Mitsutake Oshikiri, M. Boero, A. Matsushita, and Jinhua Ye, Water Molecule Dynamics at around the Co-catalyst Ni Oxide at the Surface of Metal Oxide Photo-catalyst, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabandai, Fukushima, 2011/1/20-22
- 15) Junyu Cao, Tetsuya Kako, Jinhua Ye, Fabrication of Fe-based Photo-electrode using PLD Method and its Photo-electrochemical Properties, Japan Chemical Society Annual Meeting 2011, Japan Chemical Society, University of Kanagawa, Yokohama, Kanagawa, 2011/03/26-29
- 16) Bing Yue, Tetsuya Kako, Jinhua Ye, Photocatalytic Reduction of CO<sub>2</sub> with H<sub>2</sub>O over Lamellar Niobic Solid Acid, Japan Chemical Society Annual Meeting 2011, Japan Chemical Society, University of Kanagawa, Yokohama, Kanagawa, 2011/03/26-29
- 17) Xiaoqing Chen, Tetsuya Kako, Jinhua Ye, Enhanced Incident Photon-to-current Conversion Efficiency of WO<sub>3</sub> Photonic Crystals with Inverse Opal Structure, Japan Chemical Society Annual Meeting 2011, Japan Chemical Society, University of Kanagawa, Yokohama, Kanagawa, 2011/03/26-29
- 18) Shuxin Ouyang, Jinhua Ye, AgAl<sub>1-x</sub>Ga<sub>x</sub>O<sub>2</sub> Solid-solution Photocatalysts: continuous modulation of electronic structure to enhance visible-light photoactivity

- for iso-propanol degradation, Japan Chemical Society Annual Meeting 2011, Japan Chemical Society, University of Kanagawa, Yokohama, Kanagawa, 2011/03/26-29
- 19) Ning Zhang, Shuxin Ouyang, Tetsuya Kako, Jinhua Ye, Room Temperature Synthesis of Micro/Mesoporous Zn<sub>2</sub>GeO<sub>4</sub> Photocatalyst and its Enhanced Photoreduction of CO<sub>2</sub>, Japan Chemical Society Annual Meeting 2011, Japan Chemical Society, University of Kanagawa, Yokohama, Kanagawa, 2011/03/26-29
  - 20) Tetsuya Kako, Qiuye Li, Jinhua Ye, Photocatalytic and Photophysical Properties of Titanium Oxide, Japan Chemical Society Annual Meeting 2011, Japan Chemical Society, University of Kanagawa, Yokohama, Kanagawa, 2011/03/26-29

Poster Presentation

- 21) Bing Yue, Tetsuya Kako, Jinhua Ye, Zinc Containing Carbon Nitride hybrid Materials as a Novel Photocatalysts for Hydrogen Evolution with Visible Light, 4th Int. Conf. on the Science & Tech. for Advanced Ceramics(STAC-4), Mielparque Yokohama, 2010/06/21 – 23
- 22) Junyu Cao, Naoki Kikugawa, Tetsuya Kako, Jinhua Ye, -Fe<sub>2</sub>O<sub>3</sub> electrode Photoanodic properties of pulsed-laser-deposited, 4th Int. Conf. on the Science & Tech. for Advanced Ceramics(STAC-4), Mielparque Yokohama, 2010/06/21 – 23
- 23) Xiaoqing Chen, Tetsuya Kako, Jinhua Ye, WO<sub>3</sub> photoelectrodes coupled to photonic crystals to enhance photoelectric conversion by photon-electron synergistic effect, 4th Int. Conf. on the Science & Tech. for Advanced Ceramics(STAC-4), Mielparque Yokohama, 2010/06/21 -23
- 24) Naoto Umezawa, Shuxin Ouyang, Jinhua Ye, High photocatalytic activity of Ag<sub>3</sub>PO<sub>4</sub> from first-principles calculations, The 17<sup>th</sup> Symposium of Photo Functionalized Materials Society, KSP Hall, Kanagawa Science Park, Kanagawa 2010/12/02
- 25) Yingpu Bi, Tetsuya Kako, Jinhua Ye, Facile Synthesis of One-dimensional Semiconductor Nanostructures with Enhanced Photocatalytic Properties, The 17<sup>th</sup> Symposium of Photo Functionalized Materials Society, KSP Hall, Kanagawa Science Park, Kanagawa, 2010/12/02
- 26) Guangcheng Xi, Bing Yue, Junyu Cao, Tetsuya Kako, Jinhua Ye, Fe<sub>3</sub>O<sub>4</sub>/WO<sub>3</sub> Hierarchical Core-Shell Structure: High-Performance and Recyclable Visible-Light Photocatalyst, The 17<sup>th</sup> Symposium of Photo Functionalized Materials Society, KSP Hall, Kanagawa Science Park, Kanagawa, 2010/12/02
- 27) Xiaoqing Chen, Tetsuya Kako, Jinhua Ye, Enhanced Incident Photon-Electron

Conversion Efficiency of WO<sub>3</sub> Photoanodes Based on 3D-Photonic Crystal Design, The 17<sup>th</sup> Symposium of Photo Functionalized Materials Society, KSP Hall, Kanagawa Science Park, Kanagawa, 2010/12/02

- 28) Junyu Cao, Tetsuya Kako, Naoki Kikugawa, Jinhua Ye, Photoanodic properties of pulsed-laser-deposited  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> electrode, The 17<sup>th</sup> Symposium of Photo Functionalized Materials Society, KSP Hall, Kanagawa Science Park, Kanagawa, 2010/12/02
- 29) Yuanjian Zhang, Toshiyuki Mori and Jinhua Ye, Phosphorus-Doped Polymeric Carbon Nitride: Preparation and Photocurrent-generation property, NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011, NIMS, Tsukuba, 2011/1/17-19
- 30) Yingpu Bi, Tetsuya Kako, and Jinhua Ye, Facile Synthesis of One-dimensional Semiconductor Nanostructures with Enhanced Photocatalytic Properties, NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011, NIMS, Tsukuba, 2011/1/17-19
- 31) Shuxin Ouyang, Jinhua Ye, AgAl<sub>1-x</sub>Ga<sub>x</sub>O<sub>2</sub> solid-solution photocatalysts: continuous modulation of electronic structure to enhance visible-light photoactivity for iso-propanol degradation, NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011, NIMS, Tsukuba, 2011/1/17-19
- 32) Kui Xie, Jinhua Ye, and Limitation Step of Photoelectrochemical Reduction of CO<sub>2</sub> in CO<sub>2</sub>-saturated NaHCO<sub>3</sub> solution, NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011, NIMS, Tsukuba, 2011/1/17-19
- 33) Zhiguo Yi, Jinhua Ye, and Ray L Withers, Materials Design towards Solar-driven Water Splitting, NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011, NIMS, Tsukuba, 2011/1/17-19
- 34) Junyu Cao, Tetsuya Kako, Naoki Kikugawa, Jinhua Ye, Photoelectrochemical property of Fe<sub>2</sub>O<sub>3</sub> electrode for water splitting, NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabandai, Fukushima, 2011/1/20-22
- 35) Xiaoqing Chen, Tetsuya Kako, and Jinhua Ye, Enhanced photoelectric conversion efficiency of WO<sub>3</sub> photoanodes based on photonic crystal, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabandai, Fukushima, 2011/1/20-22
- 36) Bing Yue, Tetsuya Kako, and Jinhua Ye, Photocatalytic reduction of CO<sub>2</sub> with

H<sub>2</sub>O over lamellar niobic acid, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabanndai, Fukushima, 2011/1/20-22

- 37) Ning Zhang, Shuxin Ouyang, Tetsuya Kako, Jinhua Ye, WO<sub>3</sub> nanorods with {100} facets exposed and enhanced activity in degradation of IPA, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabanndai, Fukushima, 2011/1/20-22
- 38) Peng LI, Shuxin Ouyang, Tetsuya Kako, Jinhua Ye, Artificial photosynthesis with different sacrificial agents over NaNbO<sub>3</sub>, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabanndai, Fukushima, 2011/1/20-22
- 39) Zongwei Mei, Ning Zhang, Shuxin Ouyang, Tetsuya Kako, and Jinhua Ye, Fabrication of Zinc Indium oxide/oxysulfide composite for enhanced photocatalytic H<sub>2</sub> evolution from water under visible light irradiation, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabanndai, Fukushima, 2011/1/20-22
- 40) Pakpoom Reunchan, Naoto Umezawa, Shuxin Ouyang, and Jinhua Ye, Chromium impurities in SrTiO<sub>3</sub>: A first-principles study, The 4th Japan-China Symposium on Advanced Photocatalytic Materials, Urabanndai, Fukushima, 2011/1/20-22

#### **4. International Collaborations:**

- 1) A new MEMORANDUM OF COLLABORATION was signed between the School of Materials Science and Engineering, Tianjin University, and Photocatalytic Materials Centre, NIMS, on field of “Advanced Photofunctional Materials for Solar Energy Conversion and Environment Remediation”.
- 2) A new MEMORANDUM OF COLLABORATION was signed between the Department of Chemical Engineering, University College London, and the Photocatalytic Materials Centre, NIMS, in the areas of research, education and training in the field of Renewable Energy Production.
- 3) Collaborate with Nanjing Univ., China, on “Research on solar energy conversion and environmental purification materials”. Exchanges of researchers (including graduate school students), exchanges of research information and promotion of joint research programs were actively conducted.
- 4) Collaborate with The University of Queensland, Australia, on “Research and Development of Photocatalysis Materials for CO<sub>2</sub> reduction and conversion”. Exchanges of researchers (including graduate school students), exchanges of research information and promotion of joint research programs were actively

conducted.

#### 5. Others

Organized the NIMS International Symposium on Photocatalysis and Environmental Remediation Materials 2011 (Jan.17-19, NIMS, Tsukuba, Japan), and the 4<sup>th</sup> Japan-China Joint Symposium on Advanced Photocatalytic Materials (Jan. 20-22, at the Conference Hall of Urabandai Nekoma Hotel, Fukushima, Japan)