触媒構造と反応場のナノスケールデザインによる環境調和反応の実現

（阪大院工[1]）〇山下 弘巳[1]

Nanoscale Design of Catalyst Structures and Reaction Spaces to Realize Environment-friendly Reactions

([1]*Graduate School of Engineering, Osaka University*)〇Hiromi Yamashita[1]

The utilization of unique reaction fields such as nanoporous spaces in zeolites, mesoporous silica, and metal-organic frameworks (MOFs), as well as thin film interfaces, has been applied for solid-catalyst design. By employing innovative catalyst preparation techniques, distinctive catalytic structures and surface active sites have been designed, including single-site photocatalysts (isolated metal ions and photo-functional metal complexes), ultra-fine semiconductor photocatalysts, plasmonic catalysts, non-equilibrium alloy nanoparticle catalysts, high-entropy alloy nanoparticle catalysts, and porous metal catalysts. Advanced surface structure analysis using cutting-edge spectroscopic and microscopic methods, such as synchrotron operando XAFS and aberration-corrected STEM, along with theoretical calculations, has enabled the elucidation of mechanisms and the design of new catalytic materials and reaction fields. The developed unique reaction fields and catalytic active sites have been applied to environmentally friendly reactions, including hydrogen cycle reactions (hydrogen production from water and hydrogen carrier molecules, hydrogen peroxide synthesis), carbon dioxide fixation reactions (synthesis of CO, formic acid, methanol, methane), and purification of air and water. By precisely controlling the structure of complex solid catalysts and their surrounding reaction fields at the nanoscale, and applying advanced operando spectroscopic analysis and thorough theoretical calculations, a new catalyst design method, “nanoscale design”, has been established.

Keywords：Nanoscale design, Nanocatalysts, Photocatalysts, Nanopourous, Reaction fields

ゼオライト・メソポーラスシリカ・有機金属構造体（MOF）のナノ細孔空間や薄膜界面などの特殊反応場を活用し、革新的触媒調製技術により、シングルサイト光触媒（孤立金属イオンや光機能性金属錯体）・超微粒子半導体光触媒・プラズモン触媒・非平衡合金微粒子触媒・ハイエントロピー合金微粒子触媒・多孔質金属触媒など従来と異なった特徴ある触媒構造と表面活性サイトを設計し、放射光オペランド XAFS や収差補正 STEM などの最先端分光法・顕微法を駆使した高度な表面構造解析と理論計算による機構解明を行うことで、新しい触媒材料および反応場を設計した。開発した特徴ある反応場と触媒活性サイトを活用することで、水素循環型反応（水・水素キャリア分子からの水素製造、過酸化水素合成）・二酸化炭素固定化反応（CO、ギ酸、メタノール、メタン合成）・空気と水の浄化などの環境調和型反応へ利用した。これまで不均一でとらえどころのなかった固体触媒やその周辺の反応場をナノレベルで精密に構造制御し、最先端オペランド分光解析と厳密な理論計算を適用することで活性メカニズムを解明する新しい触媒設計法（ナノスケールデザイン）を確立した。

1) H. Yamashita, K. Mori, Y. Kuwahara, T. Kamegawa, M. Che, et al., *Chem. Soc. Rev.,* **2018**, *47*, 8072.