



# 2019「中技社科技獎學金」

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## 研究獎學金 Research Scholarship



階層性沸石與介孔鈦矽酸鹽的合成與催化研究

### Synthesis and Catalytic Studies of Hierarchical Zeolites and Nanoporous Titanosilicates

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#### 研究重點

The study aims to design and prepare novel porous materials toward catalytic and other applications. Two synthetic strategies were developed to prepare distinct types of materials. Inspired by the synthesis of hierarchical zeolites comprising nanosheets using bifunctional structure-directing agents (SDAs), we developed a series of triblock SDAs featuring a polypropylene oxide (PO) linker between two heads of structure directing groups. Experimental results suggested that branching and stacking of zeolite nanosheets could be rationally controlled by using the designed triblock SDAs. Moreover, the highly-branched silicalite-1 nanosheets catalysts possessing significant amount of nest-like silanols which were generated most likely at the junctions of well-organized nanosheets/nanosheets exhibited excellent and stable activity for the vapor-phase Beckmann rearrangement and high lactam selectivity. We also employed the hydrophobic interaction between the PO linker of triblock SDA and the surface of graphene flakes to direct the restricted growth of zeolite nanosheets on graphene flakes. For the same purpose, a diblock SDA composed of a moiety of pyrene was synthesized to utilize the strong  $\pi$ - $\pi$  interaction between pyrene and graphene. We further utilized the inter-sheet space between zeolite nanosheets which served as nanoconfinement for forming the edge-attached domains of gold nanodisks (Au NDs) and titanium dioxide (TiO<sub>2</sub>) nanophases. Benefiting from the anisotropic shape of Au NDs and their edge attachment with TiO<sub>2</sub>, the plasmonic nanocomposite exhibited superior photoactivity for hydrogen production from photoreforming of methanol. Moreover, experimental results suggest that zeolite nanosheets effectively gated the access to the surface of Au NDs and allowed only methanol but not higher alcohols to react to produce hydrogen. Alternatively, we developed a surfactant-free synthesis of nanoporous titanosilicates via the homogeneous cocondensation of silicate species and triethanolamine-stabilized titanium induced by a pH-drop of the highly alkaline synthesis solution. The thus prepared materials showed high catalytic activity for simultaneous esterification and transesterification of non-edible Jatropha oil for the production of high-quality biodiesel fuel.

#### 研究成果

##### Hierarchical silicalite-1 octahedra comprising highly-branched orthogonally-stacked nanoplates as efficient catalysts for vapor-phase Beckmann rearrangement

##### Formation of hierarchical assemblies of MFI nanosheets with controlled branching by using triblock structure-directing agents

##### Hierarchically stacked MFI nanosheets with controlled stacking and inter-sheet space as robust catalysts for vapor-phase Beckmann rearrangement

##### Restricted growth of zeolite nanosheets on graphene flakes

##### Efficient hydrogen production by selective alcohol photoreforming on plasmonic photocatalyst comprising sandwiched Au nanodisks and TiO2

##### Efficient simultaneous esterification/transesterification of non-edible Jatropha oil for biodiesel fuel production by template-free synthesized nanoporous titanosilicates

#### 研究生活及心得

從事研究與追求突破是一條漫長且艱辛的路，雖然並不好走，但靠著求知慾與好奇心的驅動，使我即使面對失敗與困境總能在沈澱後繼續努力，而每每有了小突破或新發現更能夠為我的研究引擎灌注滿滿的能量。五年的付出與努力，結實了許多收穫也看到了自己的成長，這段時間雖辛苦但同時也是種享受，相當感激母校清華大學和楊家銘教授提供給我充足的資源以及對我的栽培，能夠在這樣的環境求學與做研究著實是件幸福的事。感謝中技社以及家人朋友給予我的鼓勵與支持，這些好比是一雙溫暖且強而有力的手，推動我繼續追尋夢想。願將一切收穫與榮譽與你們分享。最後期許自己今後能精益求精，更上一層，並將個人的能力奉獻給國家與社會。