論文 / 著書情報
Article / Book Information

題目(和文)	ナノ構造制御触媒を用いたメタンの二酸化炭素改質反応の低温化
Title(English)	Carbon dioxide reforming of methane under low temperature by nano-phase controlled catalyst
著者(和文)	庄司州作
Author(English)	Shusaku Shoji
出典(和文)	学位:博士(学術), 学位授与機関:東京工業大学, 報告番号:甲第11539号, 授与年月日:2020年3月26日, 学位の種別:課程博士, 審査員:宮内 雅浩,中島 章,生駒 俊之,松下 伸広,松下 祥子
Citation(English)	Degree:Doctor (Academic), Conferring organization: Tokyo Institute of Technology, Report number:甲第11539号, Conferred date:2020/3/26, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	材料 エネルギー 系 コース	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of （ 学術 ）
学生氏名： Student's Name	庄司州作	指導教員（主）： Academic Supervisor(main)	宮内雅浩
		指導教員（副）： Academic Supervisor(sub)	

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Dry reforming of methane (DRM) is attractive reaction which produce energy and reduce greenhouse gas, with converting methane (CH_4) and carbon dioxide (CO_2) into syngas (mixture gases of H_2 and CO). However, CH_4 and CO_2 are very stable, the DRM reaction requires high temperature (over 800 ˚C). Further, the high operation temperature leads aggregation of catalysts and quick carbon deposition. Thus, the new catalysis concept to drive DRM under low temperature is strongly desired.

There are two approaches for the catalyst design. First approach is the topological control of thermal catalyst to stably drive DRM. In this thesis, I developed a new class of topology-tailored catalyst in which tens-of nanometer-thick fibrous networks of Ni metal and oxygen-deficient Y_2O_3 are entangled with each other to form a rooted structure, i.e., Ni#Y_2O_3. I demonstrate that the rooted Ni#Y_2O_3 catalyst stably promotes the carbon-dioxide reforming of methane at 723 K for over 1000 h, where the performance of traditional supported catalysts such as Ni/Y_2O_3 diminishes within 100 h due to the precluded mass transport by accumulated carbon byproducts. The long-term stable methane reforming over the rooted catalyst is ultimately attributed to the topologically immobilized Ni catalysis centre and the synergistic function of the oxygen-deficient Y_2O_3 matrix, which successfully inhibits the accumulation of byproducts.

Second approach is developing photocatalyst for DRM. Photocatalytic DRM (photo-DRM) can be a promising strategy to drive DRM under extra low temperature. Previous researches of photon-driven methane conversions were the kind of photo-assisted thermal catalysis over β-Ga_2O_3, MgO, $NaTaO_3$, TiO_2, SnO_2, or WO_3-based materials, and those were usually performed at elevated temperatures while light irradiation. In other words, photocatalytic DRM using solely photons as energy source without heating has hardly been reported to date. Further, the mechanisms including charge transfer process and intermediates of photocatalytic DRM are still unclear. In this thesis, I report the nano size metal Rh and $SrTiO_3$ (Rh/$SrTiO_3$) composite, which can be synthesized by a facile hydrothermal method. The Rh/$SrTiO_3$ catalyst exhibited very high efficiency toward DRM at moderate temperature without any external heating. The conversion of gases was reached to 50 %, which was comparable to the performance of thermal catalysts driven at 500-600 ˚C Further, the *in-situ* electron spin resonance (ESR) measurement and Kelvin Prove Force Microscope analysis clarified the charge transfer process, in which the photo-excited electrons of STO by UV irradiation were injected into Rh and reacted with CO_2 as well as the photo-exited holes in STO were reacted with CH_4 to drive DRM. Further, to determine the reaction scheme and to clarify the intermediate, we conducted the isotope analysis by exchanging the lattice $^{16}O^{2-}$ of STO into $^{18}O^{2-}$. Without ^{18}O exchange condition, there was no $C^{18}O$ production, however, $C^{18}O$ molecules were produced from the ^{18}O exchanged sample, clearly indicating that the lattice O^{2-} was acted as intermediate of the reaction. This is the first report to indicate the O^{2-} ion as a mediator to drive photocatalytic uphill reaction.

Finally, I combined the concepts of topologically controlled thermal catalyst and photocatalyst to establish efficient photocatalytic DRM system on the basis of nano-structural and band-structural control. Rh meatal and lanthanoid oxide based nano-phase controlled photocatalyst efficiently exhibited photo-DRM with high conversion of 65 % and over 100 hours lifetime.

備考 ： 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。
Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).

注意：論文要旨は、東工大リサーチリポジトリ(T2R2)にてインターネット公表されますので、公表可能な範囲の内容で作成してください。
Attention: Thesis Summary will be published on Tokyo Tech Research Repository Website (T2R2).