Non-Thermal PLasma Enabled cAtalysis-Separation system for UpgRading biogas to mEthane-NTPleasure

Informe

Información del proyecto

NTPleasure

Identificador del acuerdo de subvención: 748196

Sitio web del proyecto

DOI
10.3030/748196

Proyecto cerrado

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Periodic Reporting for period 1 - NTPleasure (Non-Thermal PLasma Enabled cAtalysis-Separation system for UpgRading biogasto mEthane-NTPleasure)

Período documentado: 2018-01-15 hasta 2020-01-14

Resumen del contexto y de los objetivos generales del proyecto

Biogas is a mixture of carbon dioxide (CO2, 15~60 vol%) and methane (CH4, 40~75 vol%), which is produced by the anaerobic digestion of organic wastes such as sewage, manure, food wastes, landfill, etc. Recently, extensive efforts have been focused on the separation of CO2 from biogas to generate an enriched biomethane stream that can be used as the transport fuel in the form of Liquid Natural Gas (LNG) or Compressed Natural Gas (CNG). In addition, CO2 utilisation is also important in the mitigation of greenhouse gas emissions (albeit that from biogas the CO2 present is from the natural carbon cycle). Specifically, the conversion of CO2 into useful chemicals (e.g. methanol or CO) and fuels (e.g. CH4) has been the subject of extensive studies.
This proposal aims to develop a novel process unit that integrates the selective separation of CO2 from biogas with the transformation of the CO2 captured for the enhanced valorisation of biogas feeds. Specifically, we propose to develop an integrated membrane separation and non-thermal plasma (NTP) system that can enable the full utilisation and valorisation of biogas. In this project, we first developed a tubular SAPO-34 membrane, which has good separation performance for capturing CO2 from biogas. Thereafter, we developed several robust Ni- or Ru-based porous catalysts for efficiently catalytic CO2 hydrogenation activated by NTP to produce CH4. Finally, integrated membrane separator and NTP reactor system was developed, showing good CO2 capture and utilisation efficiency. The integrated design combining CO2 capture with CO2 methanation at ambient temperature will be an excellent candidate for further exploitation in the industrial scale biogas upgrade process.
CO2 emissions are the major contributor to the global warming and climate change as emphasised in the 2015 COP21 conference (Paris). Therefore, the fellowship project results are timely in order to tackle the challenge of CO2 capture and utilisation. Although the research under study here focused on biogas valorisation, the developed technology is generic and thus will be applicable to CO2 utilisation from both bio-based sources and fossil fuel derived emissions to reduce the impact of carbon emissions globally.

Trabajo realizado desde el comienzo del proyecto hasta el final del período abarcado por el informe y los principales resultados hasta la fecha

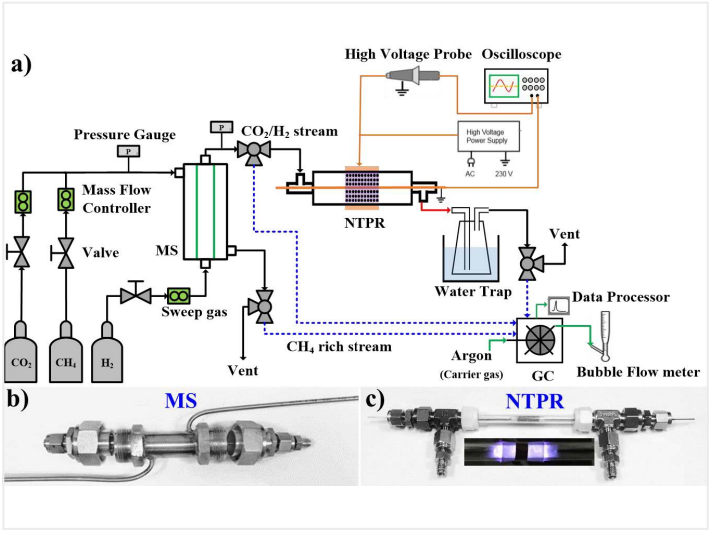
During the fellowship, work performed from the beginning of the project to the end of the period covered by the report and main results achieved so far are as follows:
(i) Fabrication of zeolite membranes with controlled thickness
During the fellowship, SAPO-34 membranes supported on tubular porous alumina were developed and applied in the capture of CO2 from biogas, showing high CO2/CH4 selectivity of 160.
(ii) Synergistic effect of NTP enabled Ni- and Ru- based catalysts systems for CO2 mathanation
For the NTP catalysis, three robust catalysts were developed, including Ni supported on zeolite (i.e. Ni/NaBETA) or metal–organic frameworks (MOFs) catalysts (i.e. 15Ni/UiO-66), Ru supported on MgAl layered double hydroxide (i.e. Ru/MgAl). These catalysts were demonstrated to efficiently enable catalytic CO2 methanation activated by NTP at ambient conditions, showing significantly higher CO2 conversions (~85 %) and CH4 yield (~84 %) at relatively low temperatures compared with

the conventional thermally activated catalysis. Additionally, the NTP activation was successfully applied to enable heterogeneous catalysis over MOFs-based catalysts for catalytic CO2 hydrogenation under mild conditions.
(iii) Catalytic CO2 methanation mechanisms under NTP activation
In situ diffuse reflectance infrared Fourier (DRIFTS) coupled with mass spectrometry (MS) characterization of the catalytic system confirmed the alternative reaction pathways enabled by NTP, enabling the fundamental understanding of the reaction mechanisms. In addition, kinetic studies of the catalytic CO2 methanation over the developed catalysts revealed that the NTP catalysis has a lower activation energy of ~21 kJ mol−1, being significantly lower than that of the thermal catalysis (~82 kJ mol−1).
(iv) process integration for improving process efficiency
Finally, the researcher carried out a proof-of-concept study of a novel integrated process, consisting of a MS followed by a NTPR in tandem, for potential application in the high-efficient capture and utilisation of CO2 towards biogas upgrading. The hybrid MS-NTPR system showed highest carbon capture efficiency (CCE) and carbon utilisation efficiency (CUE) of ca. 91.8% and 71.7%, respectively. In addition, the integrated process also exhibited excellent stability for CCU in the biogas upgrading, i.e. a stable performance over a 40 hr longevity test.

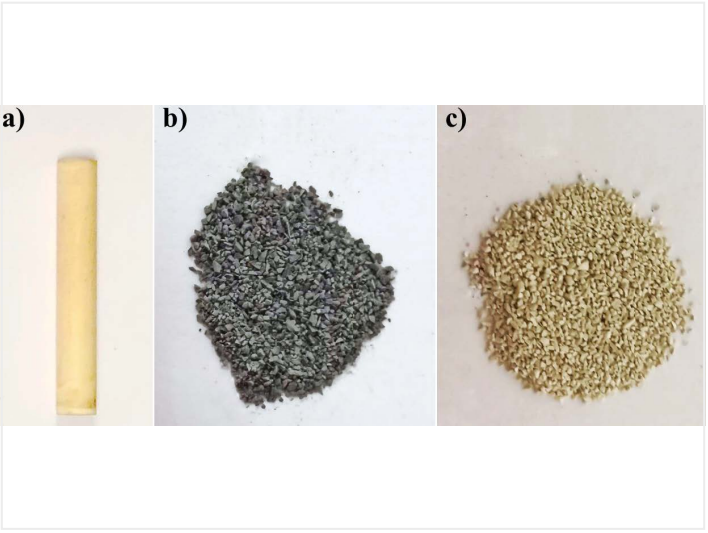
During the fellowship, the researcher has attended several conferences to disseminate the results of the project, including:
1. Chen, H.; Fan, X.; Hardacre, C. High-efficiency process for CO2 capture and utilization. Researcher Links Workshop on Sustainable Systems for CO2 Utilization in China and the UK, Beijing, China, July 2018.
2. Chen, H.; Fan, X.; Hardacre, C. Coupling of non-thermal plasma with BETA zeolite supported Ni catalysts for the methanation of carbon dioxide. UK Catalysis Conference, Loughborough, UK, January 2019.
3. Chen, H.; Fan, X.; Hardacre, C. Coupling of heterogeneous catalysts with non-thermal plasma for CO2 methanation: probing the reaction mechanisms using in-situ DRIFTS. ACS Spring 2019 National Meeting in Orlando, US, March 2019.

Avances que van más allá del estado de la técnica e impacto potencial esperado (incluida la repercusión socioeconómica y las implicaciones sociales más amplias del proyecto hasta la fecha)

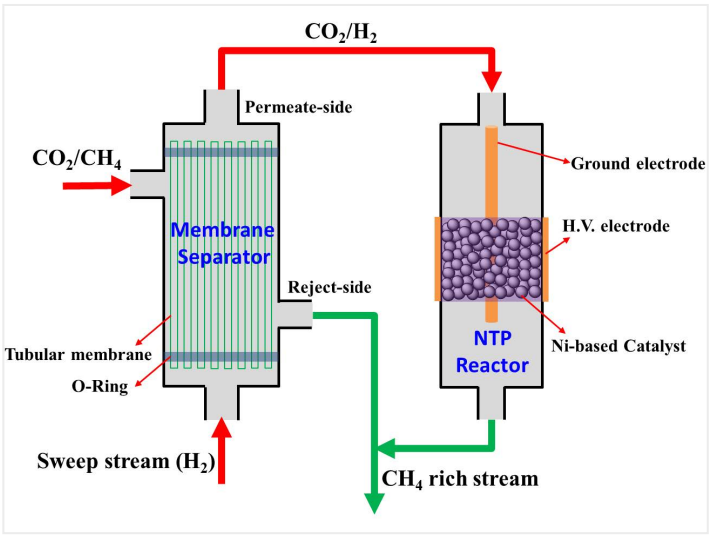
CO2 emissions are the major contributor to the global warming and climate change as emphasised in the 2015 COP21 conference (Paris). Therefore, the fellowship project results are timely in order to tackle the challenge of CO2 capture and utilisation. The research outputs have potential for further exploitation in industrial scale biogas upgrade process and CO2 utilisation.



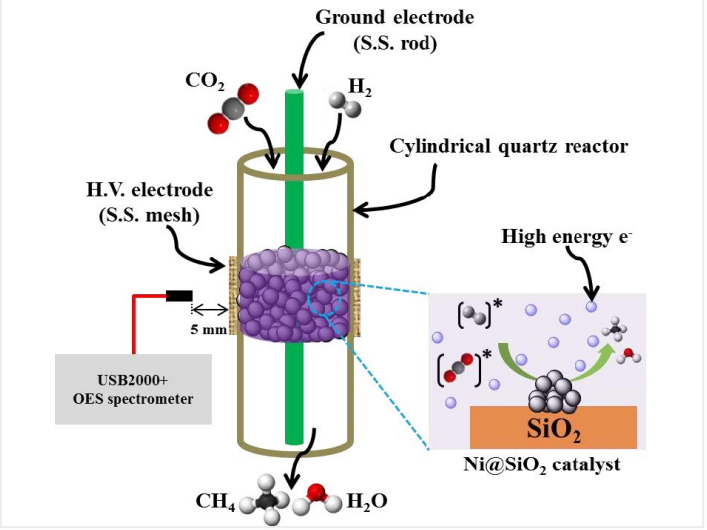
a) Lab-scale hybrid MS–NTPR experimental setup for the CO2 capture and utilisation, and photograph o



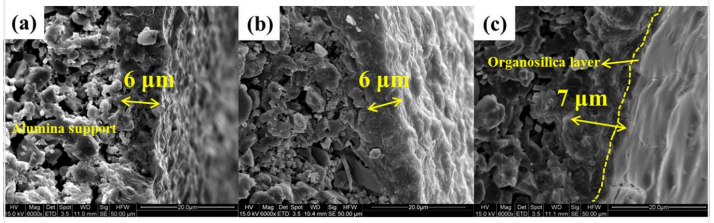
Photograph of a) SAPO-34 zeolite membrane tube, b) Ni/NaBETA, and c) Ni/UiO-66 catalysts.



Schematic of the hybrid MS–NTPR sequence configuration



Schematic representation of catalytic CO2 hydrogenation over the Ni@SiO2 catalyst in a continuous-fl



Cross-sectional SEM images of (a) VAD-OS-1-SAPO-34, (b) VAD-OS-3-SAPO-34 and (c) VAD-

OS-6-SAPO-34 me

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Permalink: https://cordis.europa.eu/project/id/748196/reporting/es