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Experimental study of mixed metal oxides catalytic materials for Dry Reforming (DR) of CH_4 and CO_2 in post-plasma use

Aim

Experimental characterization and simplified modelling of mixed metal oxides obtained from layered double hydroxides of Ni/Al/Zn/Cu/Mn in the dry reforming reaction. The catalyst is to be screened and optimized for efficient post-plasma use, accounting for its catalytic and adsorption effects.

Justification

Dry reforming (DR) of methane (CH_4) is a promising pathway to produce valuable chemicals such as carbon monoxide (CO) and hydrogen (H_2) starting from CH_4/ carbon dioxide (CO_2) mixtures. Non-thermal plasma reactors, and in particular Nanosecond-pulsed plasma discharges (NPD), have demonstrated excellent capacity to perform the reaction at reasonable energy cost. The coupling of this specific discharge with a downstream, multifunctional material offers an intriguing possibility to further enhance its capacity [1, 2]. The present study has the aim to probe and screen the performance of mixed metal oxides (MMOs), obtained from layered double hydroxides[3] (LDH) of various metals, including Ni, Al, Zn, Cu, Mn. These materials can act both as catalysts for reverse water-gas shift reaction (RWGS) and adsorbents for various molecules such as H_2O and CO_2. In this way, they offer the possibility to regulate and control the secondary undesired reactions of DR by manipulating the operating conditions. Given the current limitations of DR scalability owing partly to its selectivity, the present study proposes to investigate this matter and offer solutions in a systematic manner.

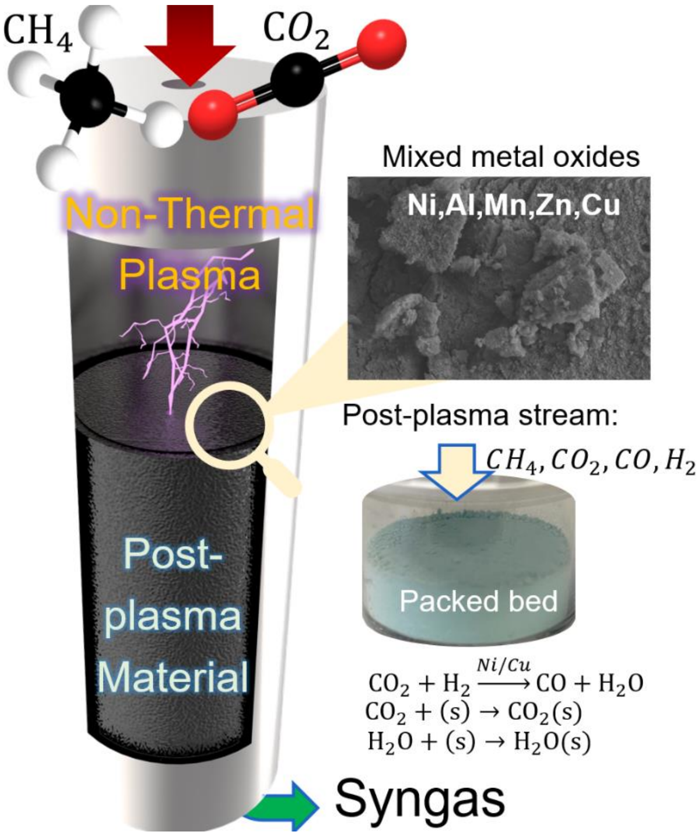

Figure 1 Schematic representation of MMOs used for post-plasma application within a packed bed configuration.

An isolated packed bed reactor will be used with variable inlet conditions, aiming to reproduce the outlet of an NPD plasma reactor. Various temperatures will be adopted together with various reactive environments (oxidative/reducing) in a step-response setup to fully probe the capacity of the material in post-plasma conditions. The limits of catalytic and adsorbent activity will be investigated and an optimal operating window will be disclosed. A 1D plug-flow pseudo-homogeneous reactor model will also be used in combination with a simplified kinetic model to describe the activity of the material. Ultimately, relevant insights will be obtained in the improvement of such materials' use in post-plasma application. The models developed will be adapted to future studies involving more sophisticated models (e.g. computational fluid dynamics reactive simulations).

Program

- Literature study on layered double hydroxides (LDH) and correspondent mixed metal oxides (MMOs), for use as adsorbent and catalytic materials.
- Characterization of the currently available MMOs (e.g. TPR, BET)
- Packed-bed material characterization experiments with previously synthesized MMOS with CH_4/CO_2 mixtures on a step-response unit.
- Kinetic model regression within the Python Cantera framework and identification of ideal post-plasma conditions.

Bibliography

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3. Veerabhadrappa, M. G.; Maroto-Valer, M. M.; Chen, Y.; Garcia, S., Layered Double Hydroxides-Based Mixed Metal Oxides: Development of Novel Structured Sorbents for CO2 Capture Applications. *ACS Applied Materials & Interfaces* **2021,** *13* (10), 11805-11813.