

Noble metal catalysts for plasma-assisted oxidation of methane

Abhinash Kumar Singh^{1*}, Irzam Javed¹, Jasmiina Palo¹, Johanna Kihlman¹, Pekka Simell¹.

¹VTT Technical Research Centre of Finland, Espoo, Finland.

*Abhinash.singh@vtt.fi

Highlights

- Plasma alone showed significant methane conversion.
- Methane conversion increased with increase in plasma power.
- Methane was mostly converted into carbon monoxide in plasma alone experiments.
- The presence of catalyst improved the selectivity of carbon dioxide.

1. Introduction

Methane (CH₄) is a potent greenhouse gas and converting methane to carbon dioxide is a promising strategy. Nevertheless, the catalytic oxidation of methane to CO₂ requires elevated temperature (above 400°C) to activate the strong carbon-hydrogen (C-H) bonds, making it energy intensive¹. Non-thermal plasma employs electrical energy to generate highly energetic electrons and reactive species, initiating diverse chemical reactions at ambient conditions, making it a suitable solution for methane oxidation². However, cold plasma itself lacks selectivity and may lead to the formation of undesirable by-products^{2,3}. Hence, the incorporation of a catalyst within the reactor can induce a synergistic effect, enhancing the selectivity of desired products³.

2. Methods

The primary objective of this study was to explore the catalytic properties of various noble metals, namely Pt, Pd, and Cu, supported on γ -Al₂O₃ and NH₄-ZSM-5, synthesized by the impregnation method. The study evaluated the oxidation efficiency of 1% CH₄ in air and the product selectivity of different catalysts under similar conditions in a Dielectric Barrier Discharge (DBD) plasma reactor. To assess the impact of plasma on the investigated catalysts, several characterization techniques were employed, including N₂ physisorption, H₂ chemisorption, XRD, and SEM.

3. Results and discussion

Plasma exhibited significant methane conversion across varying plasma power. The conversion of methane increased with increase in plasma power. The presence of catalyst reduced the methane conversion. Notably, 1% Pd/Al₂O₃ and 1% Cu/Al₂O₃ catalysts emerged as the most active, surpassing methane conversion achieved by pure γ -Al₂O₃ support. Remarkably, the presence of noble metal catalysts improved the selectivity towards CO₂. At 30 W, none of the catalysts displayed selectivity towards CO. Palladium catalysts showcased remarkable consistency with above 90% selectivity towards CO₂ across all plasma power levels. The formation of other products, including N₂O and NO_x (NO and NO₂) was observed across all plasma power levels.

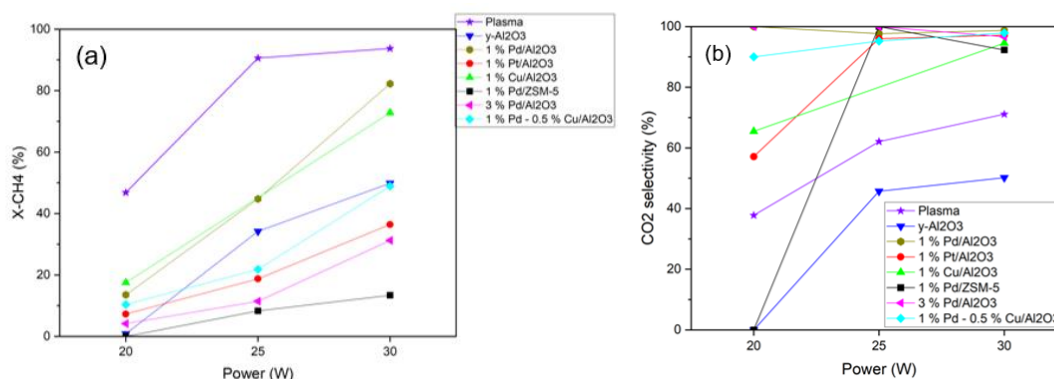


Figure 1. Effect of catalysts at different plasma power on (a) Conversion of methane (b) CO₂ selectivity.

4. Conclusions

The complete oxidation of methane can be accomplished through a plasma-catalyst system. While plasma alone demonstrates substantial methane conversion, it tends to generate undesired byproducts, such as carbon monoxide (CO). The integration of noble metal catalysts facilitates the complete oxidation of methane, enhancing the selectivity towards carbon dioxide (CO₂). The synergy between plasma and catalyst significantly elevates the selectivity for the desired product, CO₂, compared to the sole use of plasma.



Funded by
the European Union

References

- [1] P. Gelin, M. Primet, *Applied Catalysis B: Environmental*. 39 (2002) 1-37
- [2] R. Gholami, C. Stere, S. Chansai, A. Singhania, A. Goguet, P. Hinde, P. Millington, C. Hardcare, *Plasma Chemistry and Plasma Processing*. 42 (2022) 709–730
- [3] R. Snoeckx, A. Bogaerts, *Chemical Society Reviews*. 46 (2017) 5805-5863

Keywords

methane oxidation; non-thermal plasma; palladium, alumina