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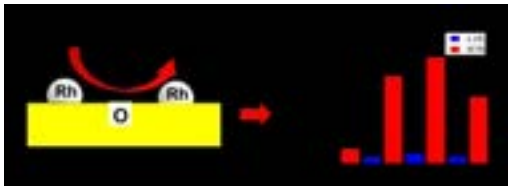
Transient CO_2 methanation over Rh/CeO_2-ZrO_2 catalysts

Statement of the Problem: The CO_2 valorization is one of the main research priorities worldwide. However, the CO_2 is a thermodynamically stable molecule that requires high-selectivity materials to react. Ceria-based catalysts have proved to be a suitable option for converting CO_2 into methane, especially the Rh/CeO_2. After applying a pretreatment of High Temperature Reduction (HTR) with pure H_2 to $1\%Rh/CeO_2$ materials, higher amounts of methane are produced during the transient state. That activity enhancement has been supposed to occur due to the additional oxygen vacancies (active sites) created after the HTR. The purpose of this study was to get new insights about the role of oxygen vacancies on the CO_2 activation, by testing materials with improved oxygen storage capacities.

Methodology & Theoretical Orientation: Polycristaline CeO_2, CeO_2-ZrO_2 and ZrO_2 supports were synthesized by precipitation/co-precipitation method. 1% Rh was then loaded by incipient wetness impregnation. The samples were characterized by means of BET surface, XRD, TPR and TGA. The produced methane after both low and high temperature reduction was followed during 1 hour.

Findings: As the introduction of zirconium into the ceria lattice enhances the reduction degree of the materials, higher quantities of methane during the transient state are produced. Therefore, a relationship between the reduction degree of the CeO_2-containing materials and the CO_2 conversion was found. However, the additional oxygen vacancies formed during the treatment are not stable at reaction conditions. Further research is needed in order to understand the mechanism for which those high-activity sites could be kept.



Recent Publications

1. Westermann A, Azambre B, Bacariza M C, Graça I, Ribeiro M F, Lopes J M and Henriques C (2017) The promoting effect of Ce in the CO_2 methanation performances on NiUSY zeolite: A FTIR in situ/operando study. Catalysis Today 283:74-81.
2. Karelovic A and Ruiz P (2013) Mechanistic study of low temperature CO_2 methanation over Rh/TiO_2 catalysts. Journal of Catalysis 301:141-153.

3. Tada S, Shimizu T, Kamayama H, Haneda T and Kikuchi R (2012) Ni/CeO_2 catalysts with high CO_2 methanation activity and high CH_4 selective at low temperatures. International Journal of Hydrogen Energy 37(7):5527-5531.
4. Novák É, Fodor K, Szailer T, Oszkó A and Erdöhelyi A (2002) CO2 hydrogenation on Th/TO2 previously reduced at different temperatures. Topics in Catalysis 20(1-4):107-117.
5. de Leitenburg C, Trovarelli A and Kašpar J (1997) A temperature-programmed and transient kinetic study of CO2 activation and methanation over CeO2-supported noble metals. Journal of Catalysis 166(1):98-107.

Biography

Monica Julieth Valencia Botero is a Project Researcher at the Vaasa Energy Business Innovation Centre (VEBIC) in the domain of the assessment of the energy systems, especially bioenergy and renewable energy. She has experience in several aspect of the energy business including technologies, environmental assessment, material science and biofuels and bioenergy. She has completed her Bachelor's degree in Chemical Engineering and MSc in Engineering-Chemical Engineering at Universidad Nacional de Colombia- Manizales, Colombia and PhD in Environmental and Energy Engineering Sciences at the University of Udine, Italy.

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