

## Effect of Water Addition on Catalytic Activity for CO<sub>2</sub> Purification from Oxyfuel Combustion

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**Abstract :** Oxyfuel combustion is a promising method that enables to obtain a CO<sub>2</sub> rich stream, with water vapor (~10%), unburned components such as CO and NO, which must be cleaned before the use of CO<sub>2</sub>. Our objective is then the final treatment of CO and NO by catalysis. Three-way catalysts are well-developed material for simultaneous conversion of NO, CO and hydrocarbons. Pt and/or Rh ensure a quasi-complete removal of NO<sub>x</sub>, CO and HC and there is also a growing interest in partly replacing Pt with less-expensive Pd. The use of alumina and ceria as support ensures, respectively, the stabilization of such species in active state and discharging or storing oxygen to control the oxidation of CO and HC and the reduction of NO<sub>x</sub>. In this work, we will compare different metals (Pd, Rh and Pt) supported on Al<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub>, for CO<sub>2</sub> purification from oxyfuel combustion. The catalyst must reduce NO by CO in an oxidizing environment, in the presence of CO<sub>2</sub> rich stream and resistant to water. In this study, Al<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> were used as support materials of the catalysts. 1wt% M/Support where M = Pd, Rh or Pt catalysts were obtained by wet impregnation on supports with a precursor of palladium [Pd(acac)<sub>2</sub>], rhodium [Rh(NO<sub>3</sub>)<sub>3</sub>] and platinum [Pt(NO<sub>2</sub>)<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>]. Materials were characterized by BET surface area, H<sub>2</sub> chemisorption, and TEM. Catalytic activity was evaluated in CO<sub>2</sub> purification which is carried out in a fixed-bed flow reactor containing 150 mg of catalyst at atmospheric pressure. The flow of the reactant gases is composed of: 20% CO<sub>2</sub>, 10% O<sub>2</sub>, 0.5% CO, 0.02% NO and 8.2% H<sub>2</sub>O (He as eluent gas) with a total flow of 200 mL.min<sup>-1</sup>, with same GHSV (2.24x10<sup>4</sup> h<sup>-1</sup>). The catalytic performances of the samples were investigated with and without water. It shows that the total oxidation of CO occurred over the different materials. This study evidenced an important effect of the nature of the metals, supports and the presence or absence of H<sub>2</sub>O during the reduction of NO by CO in oxyfuel combustions conditions. Rh based catalysts show that the addition of water has a very positive influence especially on the Rh catalyst on CeO<sub>2</sub>. Pt based catalysts keep a good activity despite the addition of water on the both supports studied. For the NO reduction, addition of water act as a poison with Pd catalysts. The interesting results of Rh based catalysts with water can be explained by a production of hydrogen through the water gas shift reaction. The produced hydrogen acts as a more effective reductant than CO for NO removal. Furthermore, in TWCs, Rh is the main component responsible for NO<sub>x</sub> reduction due to its especially high activity for NO dissociation. Moreover, cerium oxide is a promotor for WGSR.

**Keywords :** carbon dioxide, environmental chemistry, heterogeneous catalysis

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