Abatement of NO by CO on Pd Catalysts: Influence of the Support in Oxyfuel Combustion Conditions

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Abstract : The CO2 emitted from anthropic activities is perceived as a constraint in industrial activity due to taxes, stringent environmental regulations, impact on global warming... To limit these CO2 emissions, reuse of CO2 represents a promising alternative, with important applications in chemical industry and for power generation. However, CO2 valorization process requires a gas as pure as possible Oxyfuel-combustion that enables obtaining a CO2 rich stream, with water vapor (10%) is then interesting. Nevertheless to decrease the amount of the by-products found with the CO2 (especially CO and NOx which are harmful to the environment) a catalytic treatment must be applied. Nowadays three-way catalysts are well-developed material for simultaneous conversion of unburned hydrocarbons, carbon monoxide (CO) and nitrogen oxides (NOx). The use of Pd attracted considerable attention on the basis of economic factors (the high cost and scarcity of Pt and Rh). This explains the large number of studies concerning the CO-NO reaction on Pd in the recent years. In the present study, we will compare a series of Pd materials supported on different oxides for CO2 purification from the oxyfuel combustion system, by reducing NO with CO in an oxidizing environment containing CO2 rich stream and presence of 8.2% of water. Al2O3, CeO2, MgO, SiO2 and TiO2 were used as support materials of the catalysts. 1wt% Pd/Support catalysts were obtained by wet impregnation on supports with a precursor of palladium [Pd(acac)2]. The obtained samples were subsequently characterized by H2 chemisorption, BET surface area and TEM. Finally, their catalytic performances were evaluated in CO2 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% CO2, 10% O2, 0.5% CO, 0.02% NO and 8.2% H2O (He as eluent gas) with a total flow of 200mL.min-1, in the same GHSV. The catalytic performance of the Pd catalysts for CO2 purification revealed that: -The support material has a strong influence on the catalytic activity of 1wt.% Pd supported catalysts. depending of the nature of support, the Pd-based catalysts activity changes. -The highest reduction of NO with CO is obtained in the following ranking: TiO2>CeO2>Al2O3. -The supports SiO2 and MgO should be avoided for this reaction, -Total oxidation of CO occurred over different materials, -CO2 purification can reach 97%, -The presence of H2O has a positive effect on the NO reduction due to the production of the reductant H2 from WGS reaction H2O+CO \rightarrow H2+CO2

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