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A Sustainable Pt/$BaCe_{1-x-y}Zr_xGd_yO_3$ Catalyst for Dry Reforming of Methane-Derived from Recycled Primary Pt

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Abstract : Dry reforming of Methane (DRM) is considered one of the most valuable technologies for green-house gas valorization thanks to the fact that through this reaction, it is possible to obtain syngas, a mixture of H_2 and CO in an H_2/CO ratio suitable for utilization in the Fischer-Tropsch process of high value-added chemicals and fuels. Challenges of the DRM process are the reduction of costs due to the high temperature of the process and the high cost of precious metals of the catalyst, the metal particles sintering, and carbon deposition on the catalysts' surface. The aim of this study is to demonstrate the feasibility of the synthesis of catalysts using a leachate solution containing Pt coming directly from the recovery of spent diesel oxidation catalysts (DOCs) without further purification. An unusual perovskite support for DRM, the $BaCe_{1-x-y}Zr_xGd_yO_3$ (BCZG) perovskite, has been chosen as the catalyst support because of its high thermal stability and capability to produce oxygen vacancies, which suppress the carbon deposition and enhance the catalytic activity of the catalyst. BCZG perovskite has been synthesized by a sol-gel modified Pechini process and calcinated in air at 1100 °C. BCZG supports have been impregnated with a Pt-containing leachate solution of DOC, obtained by a mild hydrometallurgical recovery process, as reported elsewhere by some of the authors of this manuscript. For comparison reasons, a synthetic solution obtained by digesting commercial Pt-black powder in aqua regia was used for BCZG support impregnation. Pt nominal content was 2% in both BCZG-based catalysts formed by real and synthetic solutions. The structure and morphology of catalysts were characterized by X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). Thermogravimetric Analysis (TGA) was used to study the thermal stability of the catalyst's samples. Brunauer-Emmett-Teller (BET) analysis provided a high surface area of the catalysts. H_2-TPR (Temperature Programmed Reduction) analysis was used to study the consumption of hydrogen for reducibility, and it was associated with H_2-TPD characterization to study the dispersion of Pt on the surface of the support and calculate the number of active sites used by the precious metal. Dry reforming of methane (DRM) reaction, carried out in a fixed bed reactor, showed a high conversion efficiency of CO_2 and CH4. At 850°C, CO_2 and CH_4 conversion were close to 100% for the catalyst obtained with the aqua regia-based solution of commercial Pt-black, and ~70% (for CH_4) and ~80 % (for CO_2) in the case of real HCl-based leachate solution. H_2/CO ratios were ~0.9 and ~0.70 in the first and latter cases, respectively. As far as we know, this is the first pioneering work in which a BCGZ catalyst and a real Pt-containing leachate solution were successfully employed for DRM reaction.

Keywords : dry reforming of methane, perovskite, PGM, recycled Pt, syngas
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