Cyprus University of Technology

Faculty of Geotechnical Sciences and Environmental Management

Doctoral Dissertation

STUDY AND EVALUATION OF INNOVATIVE METHODS FOR PRINTING SOLID CATALYSTS INTENDED FOR NO_x POLLUTION CONTROL

Vasiliki K. Chatziiona

Limassol, December 2019

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DEPARTMENT OF CHEMICAL ENGINEERING

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Approval Form

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To my loving grandparents

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ABSTRACT

The present Doctoral Dissertation concerns the evaluation and development of an innovative method for preparing solid supported-Pt catalysts intended for NO_x pollution control. In particular, the alternative and cutting-edge approach of developing catalysts via multilayer inkjet printing was examined, so as to control the structure of solid catalysts at a nanoscale level, using two different printers, i.e., a modified Epson L800 printer and a commercial material printer (DMP-2850). For the first time ever, one 0.1 wt% $Pt/MgO/CeO_2$ and two 0.1 wt% Pt/Al_2O_3 catalysts were prepared by novel inkjet printing and compared, in terms of their catalytic behaviour towards the $NO/H_2/O_2$ reaction, against four catalysts prepared by a standard and a modified wet impregnation method.

It is worth mentioning that the inkjet-printed Pt/Al_2O_3 catalysts presented excellent activity and wide operating temperature window (T_R=100-250°C) towards the selective catalytic reduction of NO by H_2 under strongly oxidizing conditions (H_2-SCR) in the very low-temperature range of 100-200°C. Specifically, the Epson printed Pt/Al_2O_3 catalyst, presented X_{NO}= 91% at 150°C, while the DMP printed Pt/Al_2O_3 catalyst presented an average X_{NO}= 97% for the low-temperature range of 140-200°C and X_{NO}= 99.5% at 175°C. As for the DMP inkjet-printed $Pt/MgO/CeO_2$ catalyst, it showed remarkable catalytic performance (X_{NO}= 100%, S_{N2}= 100%, $T_R \geq$ 200°C) in the absence of oxygen (NO/H_2 reaction), a result which has never been reported before, according to the author's knowledge, in particular without the formation of NH_3 as a by-product.

Surface reactivity studies by transient methods performed within the present work indicated that the inkjet printing process leads to a unique surface structure of the printed catalysts that probably favours the formation of different intermediate NO_x species, which are active at very low reaction temperatures. Moreover, it was proven through combined SSITKA-DRIFTS studies, that the different catalyst preparation methods utilized for the development of $Pt/MgO-CeO_2$ catalysts, affects the formation and concentration of different active adsorbed intermediate NO_x species on Pt surface, as well as on the support and the metal-support interphase. Furthermore, the transient experiments revealed important information towards the understanding of basic mechanistic issues of the present catalytic system (e.g., surface coverage of NO_x intermediate species and N-containing species, H_2 spillover).

Keywords: novel catalyst inkjet printing, supported Pt catalysts, H_2-SCR, NO reduction.