Robust one-step synthesis of structural catalyst Co_3O_4|Al_2O_3|cordierite for N_2O abatement

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Highlights

- Optimization of spinel phase loading for Co_3O_4|Al_2O_3|cordierite catalyst.
- Evaluation of pressure drops for bare support, washcoated monolith and full catalyst.
- Investigation of synthesis condition impact on catalyst composition, morphology and performance.

1. Introduction

Increasing human impact on natural environment caused by dynamic industry development and increasing urbanization results in emission of hazardous substances into the atmosphere. Nitrous oxide, recognized as a one of the most dangerous greenhouse gases [1], is emitted annually in large quantities. Although agriculture sources dominate N_2O emission, they are uncontrollable in comparison to wastes (i.e., hospital installations) or industrial processes (i.e., nitric acid plant). Due to the high harmfulness of emitted gases, there was a discussion taken up in Kyoto in 1997, which covered limiting their emission. According to the currently binding European Commission Directive, all of EU members should significantly reduce their emissions as soon as possible. Therefore, researchers from around the world have been seeking an optimal solution for this problem for a long time.

Literature data analysis indicates that, in the context of N_2O decomposition, a big number of catalysts with various chemical compositions, types of system (bulk, supported) and forms (pellets, tablets, extrudes, rings) have been investigated so far. Cobalt spinel was found as one of the most stable and active phase in deN_2O reaction. The problem of high prize of bulk cobalt catalyst can be easily solved by its dispersion over support. Herein, cordierite ($2MgO$-Al_2O_3-$5SiO_2$) monolith with parallel, square channels have been chosen as a support body for cobalt spinel catalyst dedicated for deN_2O reaction in hospital installations and nitric acid plant. This commonly used support provides a good contact between the active phases of the catalyst and the gas reactants, with high mechanic resistance and dust tolerance as well as low-pressure drops.
The aim of this study was to optimize the process of cobalt active phase deposition on monolith body by solution combustion synthesis (SCS). Various parameters of preparation procedure (kind and amount of fuel, ignition temperature, cobalt spinel loading) were investigated. Moreover, the impact of both, loading and washcoating, on pressure drops was tested. Additionally, evaluation on internal and external mass transfer was carried out.

2. Methods

The series of catalysts Co_3O_4|Al_2O_3|cordierite was prepared via SCS on monolith cordierite support, primarily washcoated by α-alumina (to avoid possible ion-migration from support to active phase structure). The optimization of the synthesis conditions was carried out towards selection of kind (urea, glycine) and concentration (12-150% of stoichiometric amount) of fuel, loading of active phase (1-10 wt. %) as well as temperature of ignition (250 – 500 ºC). As prepared samples were compared by a series of screening tests in

deN$_2$O reaction (TPSR, 5% N$_2$O/He), by varying the gas hourly space velocity (2,000-10,000 h^{-1}). The external and internal mass transfer were calculated with the Carberry number and Wheeler-Weizs criterion. All synthesized catalysts were carefully analyzed by means of µRS, XRD, and SEM techniques in order to confirm the presence of cobalt spinel structure and analyze the morphology of obtained materials.

3. Results and discussion

The structural and surface analysis of obtained structures indicates the presence of cobalt in all obtained samples. Microscopy analysis confirms also that the active phase, with nanometric size of Co_3O_4 crystals, is very well dispersed on the micrometric alumina washcoat. Considering the optimization of cobalt spinel preparation conditions, glycine fuel allows to obtain more active catalyst, especially in amounts of fuel under the 50% of stoichiometric quantity. Lower temperature of ignition is completely enough for carrying out SCS, however in order to stabilize the obtained catalyst, all samples should be calcined at the highest temperature of dedicated process (600 ºC). Pressure drops caused by washcoating and active phase deposition are not significant for the laboratory scale of experiments.

4. Conclusions

The series of catalyst Co_3O_4|Al_2O_3|cordierite were obtained via dip-coating (alumina washcoat) and solution combustion synthesis (cobalt spinel deposition) on structural cordierite support. Selected conditions of cobalt spinel procedure preparation (kind and amount of fuel, ignition temperature, active phase loading) were optimized towards activity in low-temperature catalytic N_2O decomposition reaction. Solution combustion synthesis with using glycine fuel in amount under the 50% of stoichiometric quantity as well as low ignition temperature ensure obtaining samples with satisfying dispersion and favorable morphology of structures. Pressure drops quantities evaluated during various temperature experiments were rated as slightly influencial for the laboratory scale experiments.

References

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Keywords
Cobalt spinel, N_2O decomposition, structural catalyst, dip-coating

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