SELECTIVE CATALYTIC REDUCTION OF NO_X WITH AMMONIA OVER CATALYSTS DERIVED FROM FLY ASH

Agnieszka Szymaszek, Paulina Summa, Bogdan Samojeden, Monika Motak

AGH University of Science and Technology, Kraków, Poland

INTRODUCTION

The generation of adverse pollutants caused by combustion of fossil fuels is nowadays a significant environmental problem. One of the most abundant and complex anthropogenic by-product of coal combustion is fly ash (FA). The term fly ash refers to particles which are collected from flue gas by mechanical or electrostatic precipitator. The physicochemical properties of this solid waste are determined by the type of combusted coal. Inappropriate disposal of FA can result in degradation of the soil and poses great danger to the environment [1-3]. Therefore, there is a pressing need to find an effective method for fly ash utilization. Over the past few years, much attention has been also directed to NO_x abatement and the most effective current method is selective catalytic reduction with ammonia (NH_3-SCR). According to the scientific literature, supported transition metals and their oxides can be potentially used as more economical substitutes of the commercial catalyst [4-5]. In view of the above, in this study the suitability of fly ash for the synthesis of NH_3-SCR catalysts supports was investigated. The concept of this kind of modification was motivated by the fact, that contaminating fly ash could be utilized with simultaneous abatement of another adverse compound. Hence, the idea is a promising opportunity to meet the objectives of the circular economy.

EXPERIMENTS

In the research, the catalytic performance in NH_3-SCR of the fly ash-derived catalysts containing transition metal oxides was investigated. Before the modification, the coal fly ash was dried in the oven at 100°C for 24 h. Subsequently, it was treated with 1 M solution of HNO_3 with the FA/acid ratio of 1:10. The slurry was left to react at 100°C for 4 or 24 h. The mixture was continuously filtered, washed and dried at 100°C for 24 h. Transition metals were introduced on the fly ash-based support via impregnation by metal nitrates. The structure of prepared materials was characterized by XRD and FTIR, the S_{BET} and texture were determined by the low-temperature N_2 sorption. The type and aggregation state of active metal species were analyzed by UV-vis-DRS. Subsequently, the obtained samples were subjected to NH_3-SCR catalytic tests.

RESULTS AND DISCUSSION

Catalyst characterization

The physicochemical characterization of the prepared materials had a significant contribution into the understanding of their properties. The specific surface area analyzed by low temperature N_2 sorption increased significantly after the treatment with acidic medium. FTIR analysis indicated the presence of characteristic absorption bands ascribed to anions and cation-oxygen bonds, hydroxyl groups and hydroxyl groups linked to Mg-, Al- and Ca- lattice. The results of XRD analysis indicated the presence of characteristic reflections of SiO_2, Al_2O_3, CaO and Fe_2O_3. These compounds are considered to influence the catalytic activity of the material in NH_3-SCR. UV-vis-DRS analysis confirmed that active metal phase occurs in different forms, including isolated cations, metal oxides and spinels.

Catalytic tests

The results of NH_3-SCR catalytic tests for the fly ash-based catalyst containing nickel and iron are presented in Chart 1 and Chart 2. The peak conversion of about 55% is reached at 200^{o}C and stays constant until the temperature does not exceed 275^{o}C. The decrease of catalytic activity above this temperature is probably caused by oxidation of ammonia, which undesired side-reaction of NH_3-SCR. The conclusion is with agreement with the results of N_2O concentration in this temperature range. Increased amount of N_2O in flue gas from about 300^{o}C indicates that NH_3 is being oxidized.

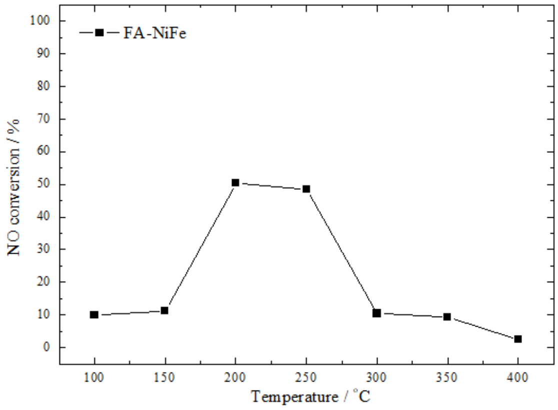

Chart 1: NO conversion for fly ash-based catalyst containing nickel and iron (reaction conditions: 800 ppm NO, 800 ppm NH_3, 3.5 % vol. O_2, balance He).

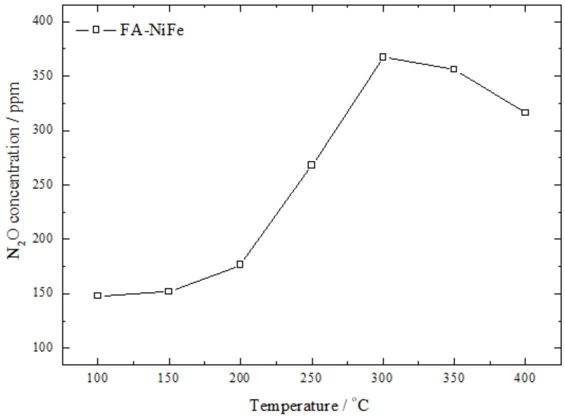

Chart 4: N_2O concentration for fly ash-based catalyst containing nickel and iron (reaction conditions: 800 ppm NO, 800 ppm NH_3, 3.5 % vol. O_2, balance He).

CONCLUSIONS

The chemical treatment of fly ash with acidic medium had significant impact on structural and catalytic properties of the material. Especially, the specific surface area was greatly increased which results in better catalytic activity in NH_3-SCR. Metal oxides present in the fly ash-based supports are predicted to enhance catalytic performance. Inclusion of active metals, e.g. iron and nickel influenced NO conversion, however further catalytic tests over materials containing copper and cobalt are predicted.

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