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Plasma and catalyst for the oxidation of NO_x

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The removal of NO_x from the exhaust gases requires the oxidation of most abundant NO to NO_2 or N_2O_5. The oxidation can be done by non-thermal plasma but the efficiency is limited due to the back-reaction of NO_2 to NO by O radicals. Present contribution investigates the role of catalysts in the improvement of oxidation efficiency based on the stationary and time-dependent studies of the NO_x oxidation at different reactor configurations and experimental conditions. The plasma produced active oxygen species (O, O_3) were shown to play an important role in the reactions taking place on the catalyst surfaces while the exact mechanism and extent of the effect depended on the reactor configuration. The effect of catalyst at different experimental conditions was quantitatively described with the aid of analytical lumped kinetic models derived for the NO_x oxidation when the catalyst was directly in contact with plasma or only with the ozone.

1. Introduction

NO_x (NO, NO_2) produced in the burning of fossil fuels is a major threat to the environment [1]. The available adsorption or absorption based NO_x removal methods work efficiently when the most abundant NO_x constituent NO is oxidized to NO_2 or N_2O_5. The oxidation can be done by non-thermal plasmas which produce highly reactive oxygen species (O, OH, O_3). One of the main limiting factor for plasma oxidation of NO in O_2:N_2 mixtures is the back-reaction of NO_2 to NO by O radicals [2]. The presence of such back-reaction distinguishes the NO_x from organic compounds which are also often oxidized by plasma. The back-reaction can be suppressed by different means including the indirect treatment by ozone or the use of catalyst.

Present contribution investigates the role of catalyst in the plasma oxidation of NO_x. The investigation involved stationary and time-dependent studies of NO_x oxidation with different configurations of plasma and catalyst placement at varying experimental conditions. Furthermore, analytical lumped kinetic models incorporating effective reaction coefficients for NO_x oxidation were derived to quantify the effect of catalyst at different experimental conditions [3,4]. The results were mostly obtained in dry O_2:N_2 mixtures.

2. Main results

Stationary and time-dependent experiments with different binary metal-oxides placed either directly in contact with plasma or after the active plasma zone demonstrated that the metal-oxides could act both as NO_x adsorbents and oxidation catalysts.

With the catalyst directly in contact with the plasma, the back-reaction in gas phase remained important but the reaction balance was shifted towards the production of NO_2. The time-dependent changes of the NO_x concentrations suggested that the surface processes involved both the adsorption/disproportionation reactions of NO_2 [5] and reactions with plasma produced oxygen species. The importance of the latter process was also emphasised by the production of NO_2 on the catalyst surfaces previously treated by oxygen discharge.

The back-reaction can be avoided when the plasma is used indirectly for the production of ozone which then reacts with NO_x. In this configuration the catalyst enhanced considerably the oxidation of NO_2 to N_2O_5 [3]. The surface processes involved the decomposition of O_3 to surface bound oxygen species which then aided the oxidation of NO_2 to NO_3. Subsequent formation of N_2O_5 on the surface resulted also in the increased NO_x adsorption ability of tested metal-oxides.

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