Regenerable sulfur traps: SO_x storage and release kinetics

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Introduction

Energy-efficient solutions for propulsion within the transportation sector are likely to involve lean, *i.e.*, oxygen excess, combustion. A possible concept to reduce the NO_x levels in the resulting (net-) lean exhausts is NO_x storage catalysis. Unfortunately, the NO_x storage catalyst (NSC) also shows high affinity towards irreversible storage of sulfur oxides (SO_x) which deactivates the NSC [1]. To prevent SO_x from reaching the NSC, one may include, so-called, sulfur traps upstream the catalyst in the aftertreatment system. In this project, different materials are evaluated as regenerable sulfur traps for which SO_x should be stored below 500°C and released above 600°C at lean conditions. Crucial parameters for SO_x storage and release processes are studied and the implications on the design of regenerable SO_x traps are discussed.

Materials and Methods

Bare and Pt containing CeO_2, Al_2O_3:MgO mixed oxide and Al_2O_3 have been prepared, characterized and investigated as potential materials for regenerable SO_x traps [2]. The samples were evaluated by lean SO_x adsorption and temperature programmed desorption (TPD) experiments using a continuous gas-flow reactor and synthetic gas compositions. Combined DRIFT spectroscopy and mass spectrometry were employed to obtain mechanistic information on the adsorption of SO_x. To further investigate the influence of Pt and the interaction between Pt and CeO_2 on the SO_x storage and release kinetics, lean SO_x adsorption and TPD experiments were carried out on different pairwise combinations of monolithic Pt/CeO_2, Pt/SiO_2, CeO_2 and SiO_2 samples (see inserts in Figure 1) [3]. In these experiments the SO_2 and SO_3 outlet concentrations were analyzed on-line with FTIR spectroscopy. The total surface area of the samples was measured by N_2 physisorption at 77K before and after SO_x exposure.

Results and Discussion

The initial flow-reactor studies singled out ceria as the most promising SO_x storage material thanks to high SO_x storage capacity and high regeneration efficiency. The storage kinetics was found to be significantly enhanced by the addition of Pt. The DRIFTS experiments revealed that lean SO_2 adsorption on CeO_2 proceeds via the formation of surface and bulk sulfates, where the latter is formed more rapidly for the Pt containing sample [2]. SO_x adsorption experiments with separated systems (*cf.* Figure 1) revealed that oxidation of SO_2 into SO_3 opens for additional, kinetically favorable, channels for SO_x adsorption in line with initial results. However, as the SO_x adsorption for the Pt/CeO_2 + SiO_2 system is much higher than for the combination Pt/SiO_2 + CeO_2, physical contact between platinum and ceria is also important for the overall SO_x adsorption process. Our contribution discusses these findings in terms of competitive adsorption, spill-over processes and type of storage sites. Additionally, the corresponding TPD experiments (not shown here) indicate that the actual storage process, i.e. via SO_2 or SO_3, affects the release process and is thus important for the regeneration efficiency.

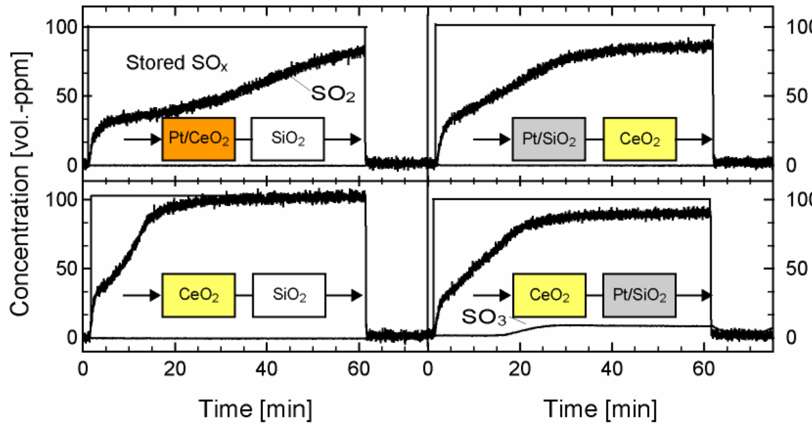

Figure 1. Responses in SO_2 and SO_3 concentrations for different pairwise combinations of monolithic samples upon lean SO_2 exposure (100 ppm SO_2 and 7% O_2 in Ar) at 250°C. Individual sample size is Ø=20 and L=18 mm and total gas flow is 3500 ml min^{-1}.

Significance

Sulfur traps can be useful to increase the durability of NSCs. Our experiments singled out Pt/CeO_2 as a promising material for regenerable SO_x traps at lean applications. On a more basic level, we have studied parameters that are important for the SO_x storage and release processes on CeO_2. The interaction of SO_x with CeO_2 is interesting not only for trap applications but for aftertreatment catalysis in general as CeO_2 is commonly used in many catalyst formulations.

References

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