Removal of formaldehyde, carbon monoxide and methane from lean gas engine exhaust gases using precious metal-free catalysts

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Significance and Relevance

Manganese and iron containing perovskite-type catalysts show high HCHO, CO and CH_4 oxidation activity in lean exhaust gases of stationary gas engines. Furthermore, the CH_4 oxidation performance was investigated at elevated pressures towards a potential pre-turbo application.

Preferred and 2nd choice for the topic: Automotive and stationary emission control / -
Preferred presentation: Oral only

Introduction and Motivations

The exhaust gas aftertreatment of lean-burn gas engines for stationary and mobile applications is of great importance for the CO_2-neutral generation of electricity and heat as well as for the mobility of the future. The engines are particularly sustainable when biogenic fuels such as biomethane are used. However, unburnt methane (CH_4) contributes to the greenhouse effect (by a factor of 25 compared to CO_2), while toxic carbon monoxide (CO) and formaldehyde (HCHO) are also emitted[1-3].

The most important catalysts for the oxidation of these pollutant are based on precious metals such as platinum and palladium, which are rare and cost-intensive. However, the catalysts are not stable with regard to the CH_4 oxidation[4]. Therefore, this paper addresses the development of novel catalysts for the removal of CH_4 as well as CO and HCHO using Fe and Mn containing perovskites.

Results and Discussion

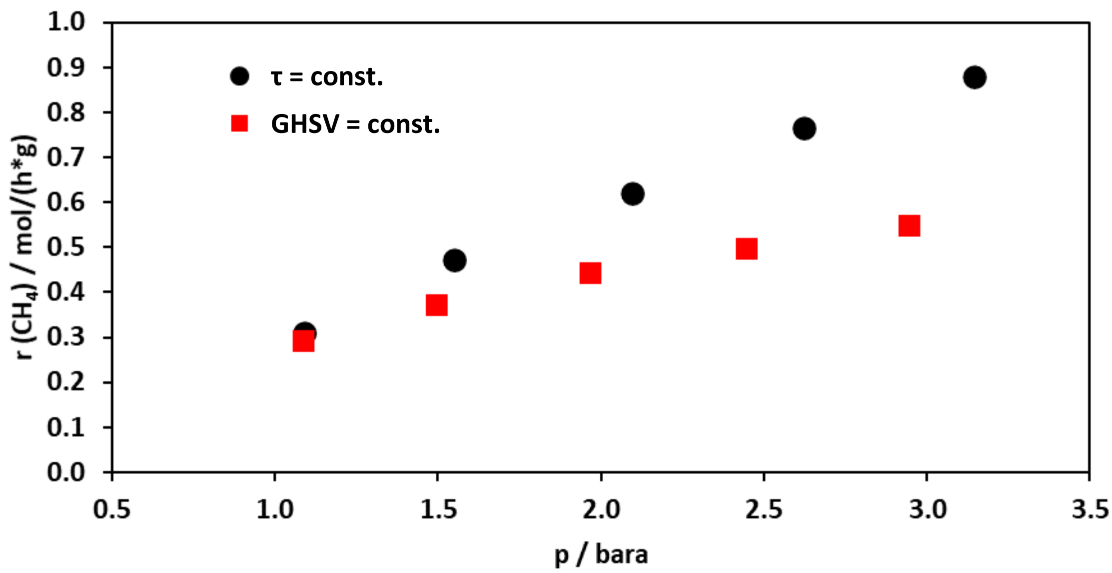

Figure 1: Reaction rate for the oxidation of CH_4 on the Mn-containing perovskite at different pressures and constant residence time (τ) or space velocity (GHSV) respectively. Conditions: y (CH_4) = 1.000 ppm, y (O_2) = 10 Vol.-%, y (H_2O) = 10 Vol.-%, N_2 balance, GHSV = 50.000 h^{-1}.

The manganese and iron containing perovskite-type catalysts reveal remarkable performance for the oxidation of HCHO and CO in synthetic as well as in the real lean exhaust gas of a stationary gas engine (not shown). By contrast, the CH_4 oxidation performance still demands significant improvement. Thus, the effect of the operation pressure on the CH_4 coverage and the resulting activity was investigated referring to a potential pre-turbo application[5]. This is illustrated in Figure 1, where an increase in pressure did indeed result in faster reaction rates for the oxidation of CH_4. As the pressure increases from atmospheric conditions up to 3 bar, both the partial pressure and the coverage of CH_4 on the catalyst rise, resulting in accelerated reaction rates. Furthermore, elevated pressure leads to prolonged residence times at the catalyst while the space velocity is maintained, which also has a beneficial impact on the conversion of CH_4. When the flow of the reactants is adjusted with increasing pressure so that the residence time at the catalyst is constant, the reaction rate is enhanced almost linearly with rising pressure.

Moreover, the effect of the pressure on the CH_4 oxidation on the Fe-based perovskite was more systematically investigated adjusting operation pressures up to 11 bar. Figure 2 demonstrates a CH_4 conversion of 95 % at 600 °C, whereas at atmospheric pressure the conversion amounts to only 50 %.

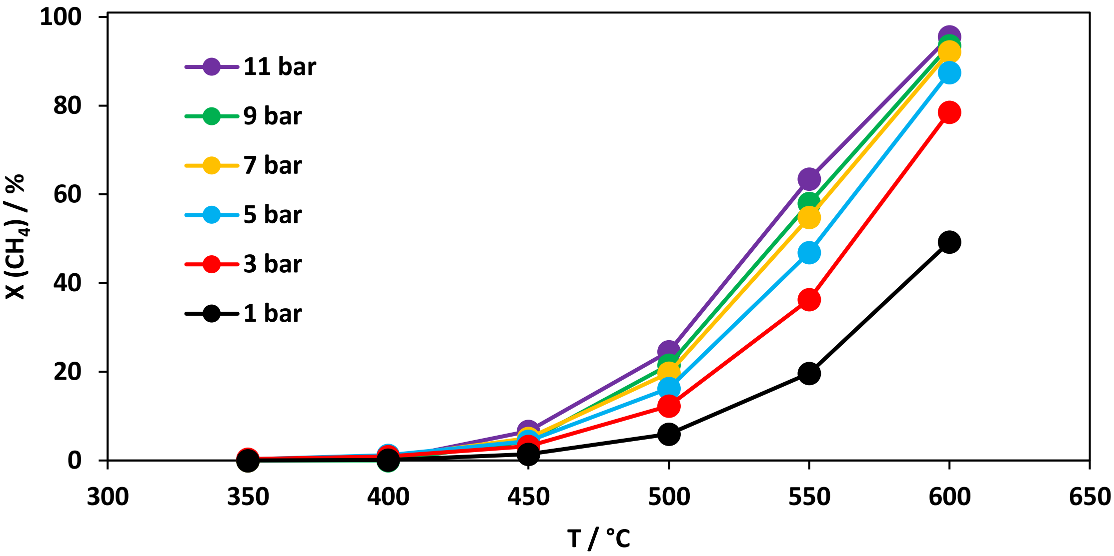

Figure 2: Conversion of CH_4 at different pressures and temperatures using the Fe-containing perovskite. Conditions: y (CH_4) = 1.000 ppm, y (O_2) = 10 Vol.-%, y (H_2O) = 10 Vol.-%, N_2 balance, GHSV = 75.000 h^{-1}.

As a consequence, the increase in pressure resulted in a drastic enhancement of the CH_4 oxidation activity suggesting that a pre-turbo application may be a promising approach to remove CH_4 from the lean exhaust of gas engines using perovskite-type catalysts [6].

References

1. S. H. Yoon, C. S. Lee, Fuel Processing Technology 92 **2011**, 5, 992–1000.
2. J. Liebetrau, J. Clemens, C. Cuhls, C. Hafermann, J. Friehe, P. Weiland, J. Daniel-Gromke, Engineering in Life Sciences 10 **2010**, 6, 595–599.
3. M. Bauer, G. Wachtmeister, MTZ worldwide 70 **2009**, 7-8, 50–57.
4. T. V. Choudhary, S. Banerjee, V. R. Choudhary, Applied Catalysis A: General 234 **2002**, 1-2, 1–23.
5. L. He, Y. Fan, J. Belletre, J. Yue, L. Luo, Renewable and Sustainable Energy Reviews 119 **2020**, article 109589.
6. M. Moser, H.-P. Walther, F. Koschany, D. Jacobsen, CIMAC Congress **2023** Busan, Paper 141.

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