



## Optimizing Iron-Containing Beta Zeolites: Influence of Synthesis Methods on Methane Oxidation Performance

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### Significance and Relevance

This study focuses on developing cost-effective, yet highly active and stable, non-precious catalysts to reduce methane emissions from marine engines, aiming to reduce greenhouse gases in the maritime sector to promote environmentally friendly maritime technologies. It also indicates a potential of iron-containing beta zeolites, already widely used for NO<sub>x</sub> reduction by SCR, as a dual-purpose application for mitigating various emissions.

*Preferred and 2<sup>nd</sup> choice for the topic: Automotive and stationary emission control, Air cleaning and combustion*

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### Introduction and Motivations

Dual fuel and gas-powered marine engines offer significant potential for reducing greenhouse gas (GHG) emissions in the maritime sector, such as CO<sub>2</sub>, NO<sub>x</sub> and particulate matter, in order to meet the growing demand for improved environmental and climate protection.<sup>1</sup> However, a key challenge remains the emission of unburned CH<sub>4</sub>. Since CH<sub>4</sub> is significantly more potent as a GHG than CO<sub>2</sub>, further oxidation to CO<sub>2</sub> is required. Until now, platinum group metal catalysts have typically been used for total oxidation of CH<sub>4</sub>. Due to their high costs and low stability against water and sulfur, novel non-precious catalysts with high activities and stabilities need to be developed. Cheap and non-toxic iron-containing beta zeolites have already been proven for selective catalytic reduction (SCR) of NO<sub>x</sub> in exhaust catalysis,<sup>2</sup> and are thus promising candidates for coupling SCR with methane oxidation. This study explores the synthesis and successful application of iron beta zeolites in the complete oxidation of methane.

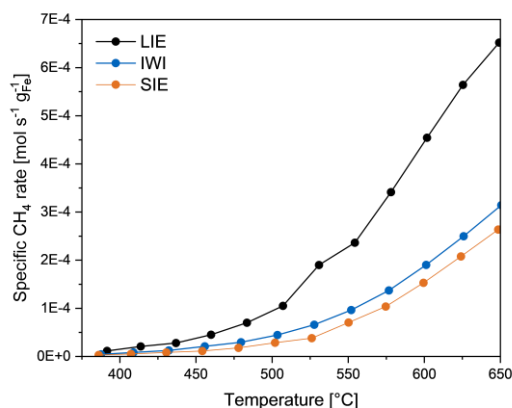
### Materials and Methods

The investigated catalysts were prepared by introducing iron nitrate into commercial beta zeolites through methods based on wetness impregnation, solid ion exchange and liquid ion exchange. As reference, the parent pure beta zeolite without iron was used. All materials were subsequently calcined at 550 °C. Structural characterization included amongst others X-ray powder diffraction (XRD), N<sub>2</sub> physisorption analysis, atomic absorption spectroscopy (AAS), transmission electron microscopy (TEM), infrared spectroscopy (IR) and temperature programmed desorption (TPD) measurements of NO. Activity measurements were conducted under specific CH<sub>4</sub> and O<sub>2</sub> concentrations at temperatures ranging from 400 – 650 °C and pressures up to 6 bar.

### Results and Discussion

Detailed characterization of the synthesized catalysts revealed that the method of iron incorporation into the zeolite influences the catalyst acidity and both the distribution and the type of iron species present. Solid ion exchange, for instance, led to the formation of a hematite phase. In contrast, no crystalline iron phase was detected in catalysts prepared by the other methods, suggesting a fine dispersion or an incorporation of iron into the zeolite framework. The results of the activity measurements are shown in Figure 1 as the specific methane conversion rate, relative to the iron loading of the catalyst. The measurements clearly indicate an impact of iron species on the activity for complete methane oxidation. Here, the catalyst from solid ion exchange (SIE) with a hematite phase

revealed the lowest activity. The catalyst from wetness impregnation (IWI) showed a higher iron dispersion and thus an enhanced activity towards methane oxidation. In contrast, liquid ion exchange (LIE) resulted in catalysts with superior activity, possibly due to better integration of iron into the zeolite framework.



**Figure 1** Catalytic activity of the catalysts depending on the temperature.

The study concludes that the method of iron incorporation critically influences catalyst properties and performance, with liquid ion exchange yielding the most active catalysts for methane oxidation due to superior iron integration into the zeolite framework.

## References

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