

Forced dynamic reactor operation for enhancing the low-temperature activity of Pd-based methane oxidation catalysts

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Palladium-based catalysts are considered most active for methane oxidation under lean conditions, but commonly suffer from pronounced water inhibition and lose their initially high activity over time.[1] In this context, forced dynamic reactor operation, i.e. short reducing pulses (SRP, absence of oxygen), is an efficient approach to boost the low-temperature activity and to overcome long-term deactivation.[2] Our recent work focuses on monolithic Pd/CeO₂ catalysts that were subject to extensive activity testing under static and dynamic conditions. Dynamic SRP operation shifts the light-off substantially towards lower temperatures and preserves high catalytic activity in humid conditions. Spatially resolved gas phase species concentration and temperature profiles along a single catalyst channel uncovered the SRP-induced formation of different zones along the catalyst (Fig. 1): A comparably low catalytic activity was found in the front zone, corresponding to less active metallic Pd covered with chemisorbed oxygen, whereas high CH₄ conversion is found in the rear part of the channel, which is attributed to the predominant presence of highly active PdO. A high support-related oxygen storage capacity helps to preserve the high activity and mitigates sintering-induced deactivation. Our work can serve as a blueprint for designing dynamic reactor operation procedures that maximize the activity of catalytic converters in general.

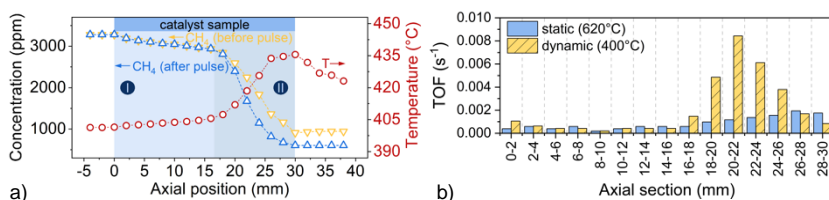


Figure 1: a) Axially resolved CH₄ concentration and temperature profile in dynamic reactor operation and b) turnover frequency (TOF) in static and dynamic reactor operation at 620°C and 400°C, respectively.[2]

[1] P. Lott *et al.*, Appl. Catal. B, 2024, **340**, 123241, DOI: 10.1016/j.apcatb.2023.123241.

[2] K. Keller *et al.*, Catal. Sci. Technol., 2024, accepted, DOI: 10.1039/D4CY00625A.