# $SO_2$ RESISTANT AND HYDROTHERMALLY STABLE HIGH SILICA Pd/CHA FOR $CH_4$ OXIDATION

**T. Mon[1], Z. Ran[2], D. Assanis[2], E. A. Kyriakidou[1,*]**

[1]Department of Chemical and Biological Engineering, University at Buffalo, The State University of New York, NY 14260, USA

[2]Department of Mechanical Engineering, Stony Brook University, NY 11794, USA

(*elenikyr@buffalo.edu)

## ABSTRACT

Methane ($CH_4$) has a 25-fold greater global warming potential than $CO_2$, limiting its use as an alternative fuel in natural gas vehicles (NGVs) due to $CH_4$ slip in the exhaust. Conventional $Pd/Al_2O_3$ catalysts used for $CH_4$ abatement in aftertreatment systems deactivate after hydrothermal aging (HTA) and $SO_2$ poisoning [1]. A promising alternative are high silica chabazite (CHA) zeolite supports that have a hydrophobic framework and are $SO_2$ resistant. Herein, high silica Pd/CHA (Si/Al = 156) are synthesized and evaluated as powdered catalysts and washcoated monoliths before and after HTA and $SO_2$ poisoning [2]. The $CH_4$ oxidation performance of powdered (100 mg, 250-500 μm, WHSV = 200 L $g_{cat}^{-1}$ $h^{-1}$) and minicore (GHSV = 20,000 $h^{-1}$) catalysts was evaluated under stoic, moderate lean, and lean conditions. The samples were pretreated in 20% $O_2$/Ar at 500 °C/20 min and were aged under stoic conditions (650 °C/ 1 h) followed by evaluation of their $CH_4$ oxidation performance. The process of aging and evaluating the catalysts was repeated for mod. lean and lean conditions. HTA was performed under 10% $H_2O$ and 20% $O_2$/Ar at 650 °C/ 12 h after which the $CH_4$ oxidation performance of the catalysts under all conditions was evaluated. The $SO_2$ tolerance (20 ppm $SO_2$/Ar, 300 °C/5 h) of HTA washcoated minicores was also evaluated [3]. Increasing the Pd loading from 1 to 2 wt.% resulted in lowering the 50 and 90% $CH_4$ conversion temperatures ($T_{50}$/$T_{90}$) to 346/373 °C (stoic), 349/386 °C (mod. lean), and 335/359 °C (lean). HTA 2 wt.% Pd/CHA powder catalyst resulted in higher $T_{50,90}$'s under stoic (352/397 °C), mod. lean (370/412 °C), and lean (353/390 °C). 2 wt.% Pd/CHA (156) washcoated minicores achieved the lowest $T_{50,90}$'s under mod. lean conditions (322/353 °C) compared to lean (322/357 °C) and stoic (348/384 °C). After HTA, deactivation was observed; however, the $T_{50,90}$'s remained below the exhaust temperatures of stoic (364/404 °C), moderate lean (337/373 °C), and lean (350/394 °C) conditions. No significant performance changes were observed over 2 wt.% Pd/CHA (156) between HTA and $SO_2$ poisoning indicating a hydrothermally stable and $SO_2$ resistant catalyst.

**KEYWORDS:** palladium, zeolites, methane oxidation, sulfur, monolith

## REFERENCES

[1] Chen, HY, Lu, J, Fedeyko, JM, & Raj, A. (2022). *Appl. Catal. A: Gen.*, 633, 118534.

[2] Prodinger S, Derewinski MA, Wang Y, Washton NM, Walter ED, Szanyi J, Gao F, Wang Y, Peden CH. (2017). *Appl. Catal. B: Environ.*, 201, 461-9.

[3] Rappé KG, DiMaggio C, Pihl JA, Theis JR, Oh SH, Fisher GB, Parks J, Easterling VG, Yang M, Stewart ML, Howden KC. (2019). Emiss. Control Sci. and Technol., 5, 183-214.