

Reduced nitrogen oxide emissions from industrial vehicles ahead

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NO_x emissions from offroad and industrial vehicles are an ongoing problem.
Credit: Shannon Colson for Pacific Northwest National Laboratory

Just a couple decades ago, nitrogen oxide emissions routinely plagued cities' skies and their residents' lungs. These polluting reactive nitrogen oxide gases from the tailpipes of combustion-engine vehicles and machinery are often abbreviated as NO_x. These emissions have, over the years, been substantially mitigated by cleaner combustion engine technologies and the implementation of exhaust aftertreatment such as selective catalytic reduction (SCR) of NO_x.

Although vehicle electrification will reduce or eliminate emissions from many mobile sources, emissions from hard-to-electrify sectors like farming and other offroad vehicles pose an ongoing challenge. What's more, achieving more efficient catalytic reduction of these emissions is becoming more difficult over time, with more efficient diesel engines producing less heat to drive the catalytic reaction. This same challenge is shared by cleaner fuels like biodiesel, or other low-carbon fuels.

Now, a new discovery by researchers at Pacific Northwest National Laboratory (PNNL), [published](#) recently in *Nature Communications*, has illuminated a previously unknown key mechanism that could inform the development of new, more effective catalysts for abating NO_x emissions from combustion engines burning diesel or low-carbon fuels.

Among best-in-class catalysts for reducing diesel emissions, a mystery

SCR of NO_x for diesel vehicles uses a reductant (typically ammonia) and a [catalyst](#) to convert NO_x to nitrogen, water, and carbon dioxide.

The researchers were comparing the efficacy of a series of best-in-class copper-based catalysts when they noticed something odd: the performance of one of the catalysts—denoted Cu/LTA—was 40% less effective at 180 °C than its counterparts, even when more reaction sites were added. The researchers couldn't explain the observation based on their prior studies.

"We wanted to understand what really caused this catalyst to be less active even though there are more [active sites](#)," said Feng Gao, a staff scientist in PNNL's Catalysis Science Group and the lead author of the *Nature Communications* paper.

The team employed electron paramagnetic resonance spectroscopy to get

a closer look at the problematic catalyst. They detected a substantial amount of the copper—meaning that it was accumulating, rather than reacting—and combined that with theoretical calculations to identify the culprit.

"Its acidity is lower than the other two," Gao explained. "Mainly, it is the lower acidity that makes the intermediate less reactive."

The researchers then used hydrothermal aging to reduce the acid sites in the other catalysts; those catalysts, in turn, showed reduced efficacy, confirming the finding.

The surprising role of acidity in diesel vehicle emissions

"A lot of the research has focused on the role of copper: how copper has to form complexes, and actually has to move around in this structure," explained Kenneth Rappe, a chief engineer and Applied Catalysis team leader at PNNL. "Then there's long been a debate as to, okay, what's the role of the acidity?"

Before this research, the researcher community had broadly understood the role of the acid sites to be storing ammonia and then providing that ammonia to the copper when needed.

"It's more than that. It actually plays an active, participating role," Rappe said. "The active copper complex that forms, in the absence of acidity, actually doesn't drive the reaction—it gets confined in space." Sans acidity, the copper accumulates rather than reacting, rendering the catalyst less effective.

The road to cleaner diesel vehicles

With this new understanding in-hand, manufacturers and researchers will be better equipped to pursue more efficient catalytic reduction of NO_x in industrial combustion engines burning diesel or low-carbon fuels.

"The acid sites are an important component to drive this reaction at low temperatures and a key consideration for designing superior catalysts that will be more active at lower temperatures," Rappe said. "It is a major development. This field has been so intensely studied. This is a significant advancement because it gives us another tool to actually improve these catalysts."

"We consider this publication a fundamental study, but this research topic is highly oriented toward applications," Gao added.

The next step for the researchers will be working with catalyst manufacturers, engine manufacturers, or both to improve the current state of the art in SCR for combustion engines burning diesel or low-carbon fuels.

"We're in the business of informing on the opportunities to design [new catalysts](#)," Rappe said.

More information: Yiqing Wu et al, Interplay between copper redox and transfer and support acidity and topology in low temperature NH₃-SCR, *Nature Communications* (2023). [DOI: 10.1038/s41467-023-38309-8](https://doi.org/10.1038/s41467-023-38309-8)

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