



Title	Development of Catalysts and Capture-Reduction Processes for N2O Abatement
Author(s)	何, 晨曦
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Development of catalysts and capture–reduction processes for N₂O abatement

(N₂O 除去に向けた分解触媒と捕集・還元プロセスの開発)

Nitrous oxide (N₂O) is a potent greenhouse gas, making its abatement a pressing environmental challenge. Thermal decomposition of N₂O offers a straightforward solution, yet this approach requires extremely high temperatures (ca. >800 °C). Currently, direct decomposition and reduction approaches are the mainstream abatement technologies with industrial applications. Between these, direct catalytic decomposition is particularly attractive owing to its efficiency and the absence of external reducing agents, which lowers operational costs and leakage risk. However, under realistic conditions, catalytic performance for direct N₂O decomposition suffers from severe deactivation in the presence of co-existing gases such as O₂. Although various materials have been explored for direct N₂O decomposition, sufficient activity is typically achieved at temperatures above 350 °C, and highly active systems in the presence of O₂ often rely on costly Rh-based catalysts. These limitations motivate the development of more active and cost-effective catalytic systems for low-temperature N₂O removal.

Machine learning (ML) has recently emerged as a powerful tool for accelerating catalyst discovery in heterogeneous catalysis. However, current approaches face two key limitations: (i) most ML studies are theory-driven, relying heavily on computational datasets such as those derived from density functional theory (DFT), which fail to capture the complexity of heterogeneous catalysis; and (ii) even ML–experiment combined studies are often restricted to narrow chemical spaces and include only one or a few closed-loop ML–experiment iterations, serving mainly as proof-of-concept demonstrations. To address these challenges, Chapter 2 introduces a ML-assisted discovery strategy using experimentally measured performance under controlled conditions and extensive closed-loop ML–experiment iterations. Starting with an initial dataset of 51 catalysts, we performed 37 cycles of the closed-loop discovery system (ML prediction + experiment) and experimentally tested 633 catalysts. Among them, more than 10 catalysts were identified to exhibit higher activity (lower T₂₀ value, the temperature required for 20% N₂O conversion during the light-off experiment) than the originally best catalyst, Rh(1)–Pd(2)/ZrO₂_RC-100 with a T₂₀ value of 378 °C. The composition of the best catalyst identified by this approach was Rh(1)–Pd(2)/ZrO₂_EP with a T₂₀ value of 355 °C.

Despite the improvements afforded by ML-identified catalysts, achieving high N₂O conversion in complex exhaust-gas mixtures still requires temperatures above 300 °C. Such high reaction temperatures not only consume large amounts of energy but also limit the applicability of these techniques. In Chapter 3, inspired by the chemical-looping strategy, we developed an N₂O capture and reduction system that uses CaO-incorporated zeolites (Ca-zeolites) as N₂O adsorbents and Pd nanoparticles on La-containing Al₂O₃ (Pd/La/Al₂O₃) as the reduction catalyst. This new process is suitable for continuous operation under temperature-swing cycling between 50 and 150 °C. The N₂O capture capacity and subsequent reduction ability were maintained for at least 15 h (10 cycles). Notably, this process can be performed at low temperatures (below 150 °C) via a simple

temperature-swing operation in the presence of O₂.

Although Ca-zeolite enables the effective N₂O capture in the presence of O₂, its performance was significantly impeded in the presence of CO₂ with the N₂O capture capacity reduced by over 90%. Moreover, selectively capturing N₂O in the presence of CO₂ remains challenging as N₂O and CO₂ share similar molecular size and physical properties. In Chapter 4, we investigated a series of materials to test N₂O adsorption and found that Cu oxides supported on CHA-type zeolite (Cu/CHA) is an efficient N₂O adsorbent in environments containing both O₂ and CO₂. Using the Cu/CHA adsorbent, we developed an N₂O capture and reduction system that could operate effectively in the presence of O₂ and CO₂ at low temperatures (below 150 °C) for at least 12 h.