Highlight

Carbon Future

Highly paired dual-atom catalysts boost ammonia electrosynthesis

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Dual-atom catalysts (DACs) have emerged as powerful platforms for complex electrocatalysis by enabling metal–metal cooperativity and multi-electron pathways that are inaccessible to conventional single-atom catalysts[1, 2]. Yet, controlling the spatial arrangement of two metal atoms, thereby achieving high pairing ratios, defined interatomic distances, and durable stabilization, remains a major challenge[3, 4]. Most confinement or sequential loading strategies produce statistically distributed dual species or aggregated nanoparticles, making it difficult to correlate atomic structure with catalytic function.

In *Nature Materials*, Wu and co-workers reported a ligand-restricted strategy that addresses these challenges by using diamine ligands to co-coordinate metal precursors and restrict their relative motion during immobilization (**Fig. 1**a)[5]. The ligands pre-organize metal ions before deposition onto two-dimensional graphitic carbon nitride (g-C_3N_4), enabling thermally stable homo- and heteronuclear architectures such as Pt–Pt and Pt–Au DACs. Unlike high-temperature precursor pyrolysis, which easily disrupts the atomic dispersion, the diamine-assisted construction preserves atomic proximity and avoids sintering, establishing a programmable chemistry for constructing atomically paired configurations on two-dimensional (2D) carbon frameworks.

Direct atomic visualization by aberration-corrected high-angle annular dark-field scanning transmission electron microscopy (AC-HAADF-STEM) confirms well-defined dual-atom motifs with long-range distribution across the g-C_3N_4 surface (Fig. 1b). The bright paired contrast patterns, supported by three-dimensional fitting and *Z*-contrast differentiation, unambiguously identify heteronuclear Pt–Au atomic pairs rather than coincidental proximity. These measurements provide rigorous structural proof and reveal that the dual-atom sites are genuinely anchored and spatially organized at the atomic scale. Such direct visual evidence is essential for correlating geometric configuration with catalytic reactivity.

A key quantitative advance is the achievement of pairing ratios exceeding 82% based on thousands of counted atoms (Fig. 1c), outperforming previously reported DAC systems prepared by space confinement, multistep deposition, or atomic layer deposition (ALD). Crucially, this high pairing ratio originates from ligand-restricted coordination rather than statistical accumulation. Moreover, tuning the diamine chain length modulates the Pt–M distance between 0.81 and 0.22 nm, enabling the synthetic control of the metal–metal coupling and electron communication that fundamentally influence intermediate binding energies and kinetic barriers. These highly paired architectures yield striking catalytic enhancement in the nitrate electroreduction to ammonia. The Pt_1-Au_1/g-C_3N_4-HA DAC achieves 744 $mg \cdot h^{-1} \cdot mg_{metal}^{-1}$ (3347 $\mu g \cdot h^{-1} \cdot mg^{-1}$) and 95.6% Faradaic efficiency (FE), representing nearly a threefold improvement relative to the unpaired Pt counterpart (Fig. 1d). This superior activity cannot be attributed solely to the metal loading or dispersion. Instead, the enhanced performance arises from synergistic dual-site mechanisms that modulate the nitrate adsorption, stabilize intermediates, and accelerate the proton-coupled electron transfer. The results demonstrate the decisive catalytic role of dual active centers and the importance of precisely engineering their spatial relationship.

The 2D g-C_3N_4 support is indispensable for realizing such atomically paired configurations. Its N-rich coordination, planar geometry, and localized electronic environment enable strong metal anchoring, contrasting sharply with curved carbon nanotubes that promote the aggregation. This illustrates a broader direction in carbon-based catalysis: Carbon frameworks increasingly serve not merely as passive hosts, but as active electronic matrices capable of regulating the charge distribution, coordination geometry, and multi-site interactions at the atomic scale. Collectively, these results establish ligand-restricted DACs as a platform for programmable atomic engineering rather than purely dispersive synthesis. From a sustainability perspective, dual-atom configurations maximize noble-metal utilization, enable multi-electron catalytic pathways, and reduce energy input, features aligned with green nitrogen-cycle chemistry, efficient ammonia synthesis, and electrochemical waste valorization. More generally, programmable dual-atom geometries constitute a conceptual interface between atomic-precision catalysis and carbon-enabled sustainable energy systems.

Beyond nitrate-to-ammonia (NO_3RR) conversion, DACs prepared on conductive 2D carbon hosts present an ideal platform for broader electrochemical technologies, including photoelectrochemical conversion and membrane-electrode assemblies. Their compatibility with delocalized π-conjugated networks could enable efficient charge transport and direct

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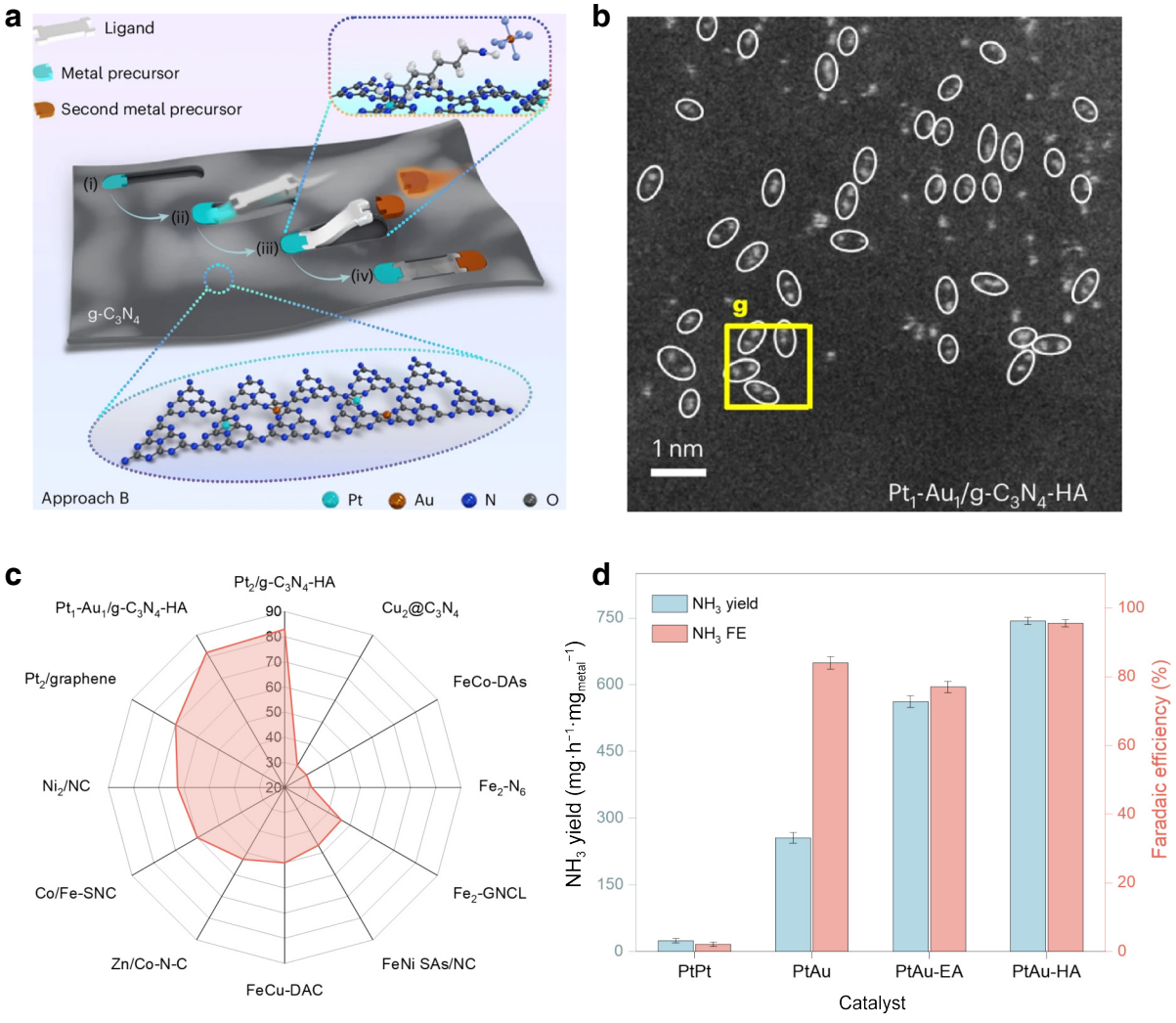

Fig. 1. Ligand-restricted construction enables programmable dual-atom platforms. a, Ligand-assisted pairing on 2D g-C_3N_4 enables controlled homo-/heteronuclear DAC formation with programmable metal–metal distance. **b,** AC-HAADF-STEM confirms the presence of true atomic pairs. **c,** Quantitative analysis shows record-high pairing ratios (up to > 82%), outperforming previously reported DAC systems. **d,** The paired Pt_1–Au_1 DAC delivers significantly enhanced nitrate-to-ammonia conversion relative to unpaired analogues, highlighting the importance of dual-site cooperativity and atomic geometrical control. Reproduced with permission from Ref[5], © Ma, Y. F. et al., under exclusive licence to Springer Nature Limited 2025.

electronic communication with electrodes, thereby minimizing interfacial losses and facilitating spatially programmed reaction environments. Coupling programmable DACs with semiconductive 2D carbon frameworks may also support light–matter interaction, promoting hybrid photocatalytic functionalities in CO_2 photoreduction or N_2 fixation under renewable operation conditions. As such, DAC–carbon architectures hold promise as integrated catalytic units bridging electrocatalysis, photocatalysis, and reactive interfaces in future sustainable energy systems: (i) Distance-dependent reaction mapping: correlating tunable interatomic separations with reaction-pathway switching supported by *operando* spectroscopy and theory. (ii) CO_2 electroreduction: engineering bimetallic distances to stabilize key *CO, *CHO, and *CO–CO intermediates toward selective C_2^+ products[6–8]. (iii) Nitrogen-cycle catalysis: exploiting dual-center cooperativity for nitrate, nitrite, or even N_2 conversion under mild conditions. (iv) Carbon-anchored multi-site networks: arranging DACs into ordered patterns to build catalytic architectures analogous to artificial metalloenzyme clusters[9]. (v) Scalable deposition: adapting ligand-restricted synthesis to continuous-flow or aerosol processes for industrially relevant catalyst production.

Overall, ligand-restricted DACs exemplify an emerging synthetic concept that unites atomic precision with carbon-mediated catalysis, establishing programmable active sites capable of addressing multi-electron transformations relevant to a low-carbon future.

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Data availability

Not applicable.

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L. L. and X. L. F. jointly wrote and revised the manuscript.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

Use of AI statement

None.