

Recent Advances and Fundamental Challenges in Computational Modeling of Electrocatalytic Ammonia Oxidation

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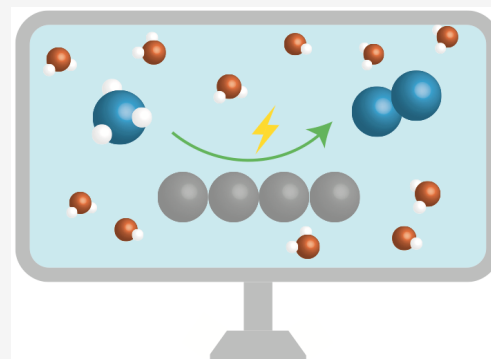
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ABSTRACT: Ammonia (NH_3) oxidation is central to the global nitrogen cycle, a delicate natural system that is now disrupted by human activities. The electrocatalytic ammonia oxidation reaction (AOR) to dinitrogen (N_2) presents a promising avenue not only for the green remediation of wastewater but also as a sustainable energy vector for the future. In this Perspective, we delve into the intricacies of AOR, highlighting the unique properties of platinum (Pt) as the best elemental metal catalyst, albeit with high overpotential and rapid deactivation. Computational chemistry as a powerful tool has provided deep insights into the nature of active sites and the elementary reaction steps of electrochemical NH_3 oxidation. We describe the structure sensitivity of this reaction with (100)-type site motifs favorable for N–N bond formation via dimerization while also touching upon the role of adsorbed hydroxyl species in dehydrogenation pathways. Addressing surface deactivation is emphasized as paramount for designing improved catalytic materials. This Perspective presents a holistic view of the recent advances, challenges, and future opportunities in computational modeling of AOR or interfacial charge transfer reactions in general, providing a roadmap for future research and innovations.

KEYWORDS: ammonia oxidation, electrocatalysis, heterogeneous catalysis, computational chemistry



INTRODUCTION

Ammonia (NH_3) oxidation represents a series of redox reactions pivotal to the global nitrogen cycle, which circulates nitrogen through Earth's atmosphere, soil, plants, and animals.¹ In this cycle, bacteria convert air's dinitrogen (N_2) into biologically usable forms and ultimately break down excessive reactive nitrogen species, channeling N_2 back into the atmosphere. It is a beautiful exchange in nature. However, humans have been tipping this delicate balance over the past century. We are burning fossil fuels, emitting huge amounts of nitrogen/sulfur oxides that lead to acid rain and smog. The Haber–Bosch process artificially fixes nitrogen in NH_3 on a massive scale of ~180 million tonnes annually. While this addresses the food demands of our expanding population, overfertilization in agriculture causes nutrient overloads in ecosystems, resulting in water pollution linked to toxic algal blooms, infant methemoglobinemia, and increased human cancer risks. Industrial and domestic wastewater containing various nitrogenous compounds such as nitrate (NO_3^-), nitrite (NO_2^-), and NH_3 further disrupt the cycle. We are stretching this natural cycle to its limits, making a global problem that needs immediate solutions (Figure 1).

The electrochemical NH_3 oxidation reaction (AOR) is of growing interest as a greener alternative to the current wastewater remediation methods, including ion exchange,

biosorption, wet air oxidation, biofiltration, and biological nitrification and denitrification.² Because NH_3 has a high volumetric energy density, low explosion risk, and well-established transportation infrastructure in comparison to hydrogen (H_2),³ NH_3 is also seen as a hydrogen-rich, energy dense fuel that can be directly used in anion-exchange membrane fuel cells (AEMFCs) or as an onboard carrier of ultrapure H_2 for proton-exchange membrane fuel cells (PEMFCs).⁴ As our climate undergoes significant changes due to anthropogenic carbon emissions, fuel cell technologies using green NH_3 present an opportunity for carbon-free, on-demand electricity.^{5,6}

In recent decades, computational chemistry has emerged as a powerful tool for understanding the mechanisms of catalytic reactions at solid surfaces and guiding the design of improved catalysts.^{7,8} For electrochemical AOR in particular, transition metal catalysts are of fundamental interest due to their promising performance for this chemistry.^{9–11} Platinum (Pt) is

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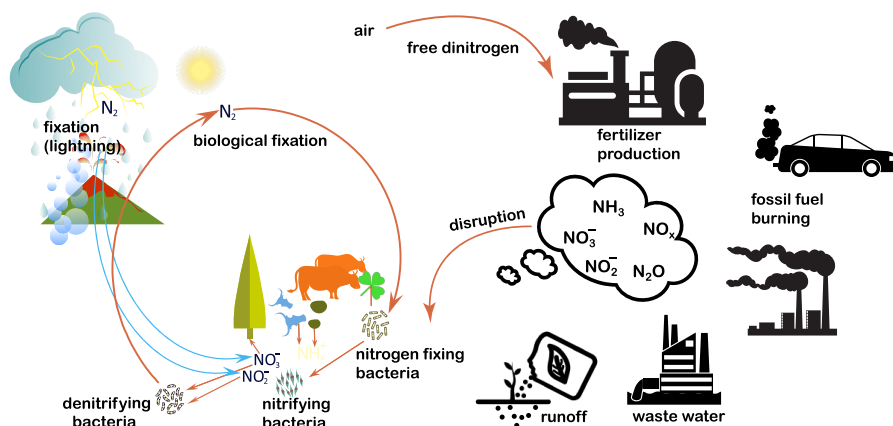


Figure 1. The global nitrogen cycle disrupted by human activities.

the best elemental metal catalyst for this reaction,¹² followed by Ir, with Co, Pd, and Rh being considered inactive^{13,14} from experiments. In a computational study by Herron et al.,¹⁵ the activities were ranked as Pt > Ir > Cu > Pd > Rh > Co. This trend was obtained from the Sabatier analysis based on activation energies and thermochemistry from density functional theory (DFT) calculations at (111) metal surfaces. For Rh, Co, Ir, Pt, and Pd, the rate-determining step is N–N bond formation, and Pt has the most appropriate nitrogen binding strength. The activity of Cu is limited by the proton–electron transfer steps required to activate NH₃ and is slightly lower than that of Ir. One of the primary reasons for Pt's superior activity in the AOR is its unique electronic structure. Pt possesses an optimal *d*-band center that facilitates the efficient adsorption and desorption of the reaction intermediates. This balance ensures that the intermediates do not bind either too weakly (leading to reduced activity) or too strongly (resulting in catalyst poisoning). However, Pt suffers from high overpotential and fast deactivation.¹⁰ It is widely accepted that (100)-type surface sites are predominantly more active than those on (111) and (110) facets.^{16–19} Alloying Pt with Ir reduces the onset potential of NH₃ activation.^{20,21} Nevertheless, for large-scale adoption of this technology, it is important to develop catalysts with minimal precious metals and improved stability while efficiently catalyzing the oxidation of NH₃ to N₂.

In this Perspective, we first cover the basics of nitrogen chemistry, providing a reference point for the electrochemical transformations of nitrogen species within a broad context. We then discuss recent advances in the computational modeling of NH₃ oxidation to N₂ on Pt, benchmarking with experimental measurements if available. We focus on the nature of active sites and elementary reaction steps occurring therein, along with design principles of new catalytic materials. We also discuss the challenges and opportunities in computationally modeling electrochemical NH₃ oxidation toward sustainable energy and environmental technologies.

■ BASICS OF ELECTROCHEMICAL TRANSFORMATIONS OF NITROGEN SPECIES

Electrochemical transformations of nitrogen species involve the interconversion of ammonia (NH₃), hydrazine (N₂H₄), hydroxylamine (NH₂OH), dinitrogen (N₂), nitrogen oxide (NO), nitrite (NO₂[−]), and nitrate (NO₃[−]) through a series of elementary reaction steps.^{22,23} We include the equilibrium

potentials of relevant nitrogen chemistry in alkaline conditions (pH = 14) on the reversible hydrogen electrode (RHE) scale (Figure 2), derived from the experimental free energy change

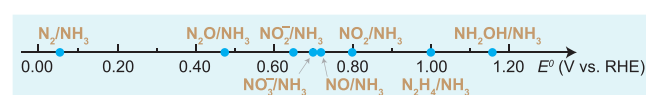
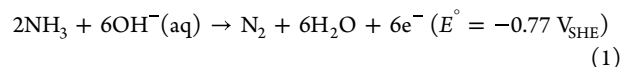


Figure 2. A redox potential diagram of electrochemical transformations of nitrogen species at pH = 14.^{24,26–28}

(ΔG) of a reaction that is a measure of the spontaneity of the transformation. It is related to the standard free energy change (ΔG°) by the equation $\Delta G = \Delta G^\circ + RT \ln(Q)$, where Q is the reaction quotient, R is the gas constant, and T is temperature. ΔG° can also be calculated using quantum chemistry and statistical mechanics, as it is fundamentally linked to the equilibrium constant K when ΔG is zero, i.e., $\Delta G^\circ = -RT \ln(K)$. The standard potential (E°) of a redox reaction is related to ΔG° of a reduction process by $\Delta G^\circ = -nFE^\circ$, where n is the number of transferred electrons, and F is Faraday's constant. For example, the standard Gibbs free energy change of NH₃ oxidation to N₂ in alkaline conditions at 25 °C (as written in eq 1) is −4.62 eV. The standard equilibrium potential for the corresponding reduction process would be −0.77 V vs SHE (standard hydrogen electrode), which is $E^\circ = +0.05$ V vs RHE (reversible hydrogen electrode).²⁴



However, in practice, it requires an extra potential, known as overpotential, beyond what thermodynamics requires to attain any appreciable current densities.²⁵ The thermodynamics of electrochemical NH₃ oxidation provide important guidance on the feasibility of the process. As shown in Figure 2, within the potential window of 0.056–0.48 V vs RHE, there is no other favorable product other than N₂ for AOR, ensuring its selectivity in low operating potentials of direct NH₃ fuel cells. This thermodynamic principle can be used to guide the design of high-performance electrochemical systems by leveraging specific NH₃ chemistry.

■ COMPUTATIONAL MODELING OF ELECTROCHEMICAL AMMONIA OXIDATION

In recent years, computational modeling has helped us tremendously in gaining insights into reaction mechanisms,

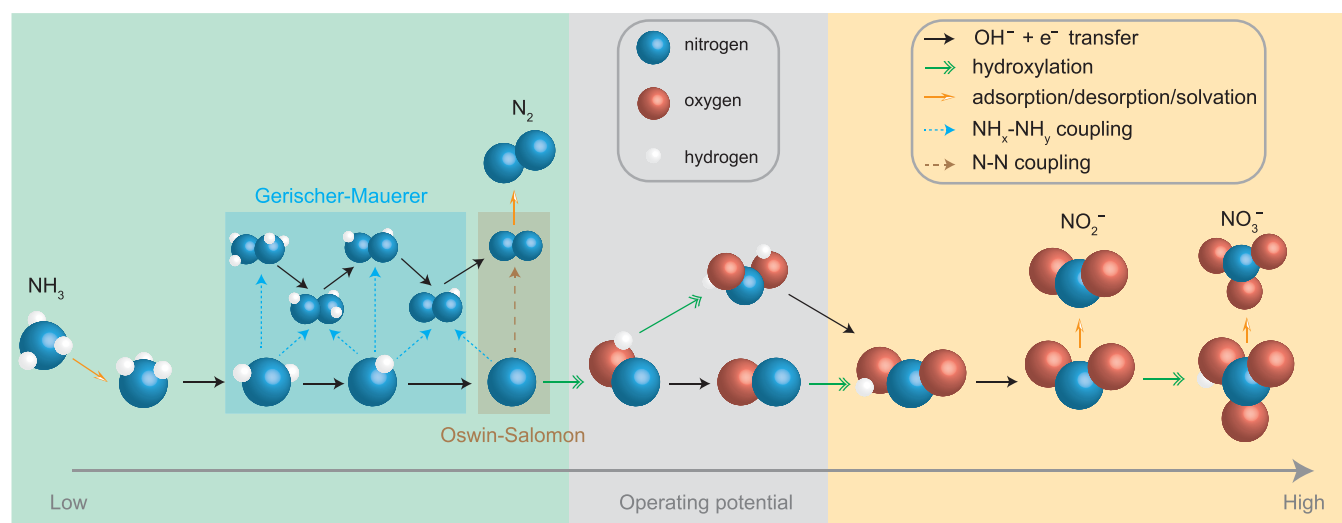


Figure 3. Reaction pathways of ammonia oxidation to N_2 with three potential regions for N_2 generation, poisoning by $*NO$, and NO_x formation. Adapted from ref 38 with permission. Copyright American Chemical Society.

structure sensitivity, the role of adsorbed hydroxyl species, and the surface deactivation of NH_3 oxidation on metal surfaces, specifically Pt. Although those aspects are intertwined in nature, we discuss them separately in the following sections.

Reaction Mechanisms. Computational modeling has played a crucial role in advancing our understanding of AOR mechanisms on metal surfaces, particularly for the best elemental metal catalyst, Pt. Two general mechanisms have been proposed within the past decades (Figure 3): the Oswin–Salomon (O–S) mechanism²⁹ and the Gerischer–Mauerer (G–M) mechanism.³⁰ The O–S mechanism was proposed in 1963, according to which NH_3 is adsorbed at surface sites ($*$) and sequentially dehydrogenated to $*N$ by OH^- ions; then, two $*N$ adatoms subsequently dimerize on surface sites to form N_2 . The G–M mechanism was originally proposed in 1970, in which NH_3 can be partially dehydrogenated to $*NH_x$ ($x = 1$ or 2) by $*OH$; then, the coupling of $*NH_x$ species takes place to form $*N_2H_y$, e.g., adsorbed hydrazine ($*N_2H_4$), followed by the stepwise dehydrogenation of $*N_2H_y$ to N_2 . It is important to note that neither of these mechanisms include pathways for NO generation, which may occur concurrently with N_2 generation at high potentials prior to the possible formation of nitrite (NO_2^-) and nitrate (NO_3^-)¹⁰ (Figure 2). The stage at which N–N bonds emerge is what differentiates the O–S and G–M mechanisms (Figure 3). The debate between $*OH$ or OH^- (aq) as the reactive proton acceptors will be discussed later in the **Role of Adsorbed Hydroxyl Species** section. In the O–S mechanism, N–N bonds form from $*N$ species, while in the G–M mechanism, the partially hydrogenated $*NH_x$ species are dimerized and $*N$ is instead considered as a poisoning species. In our opinion, this is a subtle difference. There is evidence that the AOR mechanism on Pt is potential-dependent with the G–M mechanism prevailing at low potentials, under which full dehydrogenation to $*N$ is prohibited, and the O–S mechanism being favored at moderate to high potentials.⁹ On Pt(111), NH_3 dehydrogenation to $*NH_2$ limits its onset potential. Once generated, the dimerization of $*NH_2$ is kinetically possible with a barrier of 1.02 eV,¹⁵ albeit severely limiting the kinetic current.¹⁵ This is consistent with the in situ infrared (IR) observation of the NH_2 wagging mode in $*N_2H_4$ on the Pt thin-film electrode,³¹

mainly populated with (111)-type surface sites. On Pt(100),¹⁰ constant-potential DFT calculations that explicitly consider the coadsorption of $*OH$ at a nearby site and include implicit solvation at the solid–electrolyte interface suggest that the dehydrogenation pathway is open toward $*NH$ at an onset potential of ~ 0.5 V vs RHE.^{9,10} The bridge-stabilized $*NH$ species can dimerize to form $*N_2H_2$ with a surmountable barrier of <0.5 eV. This theoretical insight aligns well with the measured onset potential of NH_3 oxidation.⁹ Even within the G–M mechanism, the true dimerization species on Pt(100) is still under debate.^{9,10,32,33} At 1/2 ML, $*NH_2$ species prefers the Pt–Pt bridge site of Pt(100), from which the dimerization barrier is significantly high (>2 eV); however, at an extremely high surface coverage,³³ $*NH_2$ favors the Pt atop site, where it can dimerize easily with a barrier of 0.3 eV at 1 ML coverage.³³ Incorporating adsorbate–adsorbate interactions into micro-kinetic modeling,¹¹ coupled with in situ spectroscopic measurements, is essential for probing the self-consistent coverage of surface species and resolving the mechanism of elementary reaction steps. For a more accurate modeling of adsorbate coverage at operating conditions, going beyond a mean-field treatment might be necessary to consider adsorbate–adsorbate interactions. One of the common approaches is to solve a stochastic Markov process within a lattice-based kinetic Monte Carlo (KMC) framework.³⁴ KMC has been extensively employed in kinetics modeling in surface science, for example, to account for local coverage effects,³⁵ simulate surface diffusion processes,³⁶ and model crystal growth.³⁷

Structure Sensitivity. One of the key factors that affects the electrocatalytic AOR activity of metal surfaces is structure sensitivity, which refers to the dependence of the reaction rate on the crystallographic orientation and surface structure. First-principles DFT calculations have been widely used to investigate the structure sensitivity of electrochemical ammonia oxidation on metal surfaces.^{9,10,15,17} These computational models have provided valuable insights into the underlying origin of the structure sensitivity. It has been shown from experimental single-crystal electrode studies that Pt(100) is more active than the Pt(111) and Pt(110) sites in terms of peak current densities (Figure 4a).^{16,19} (111)-type

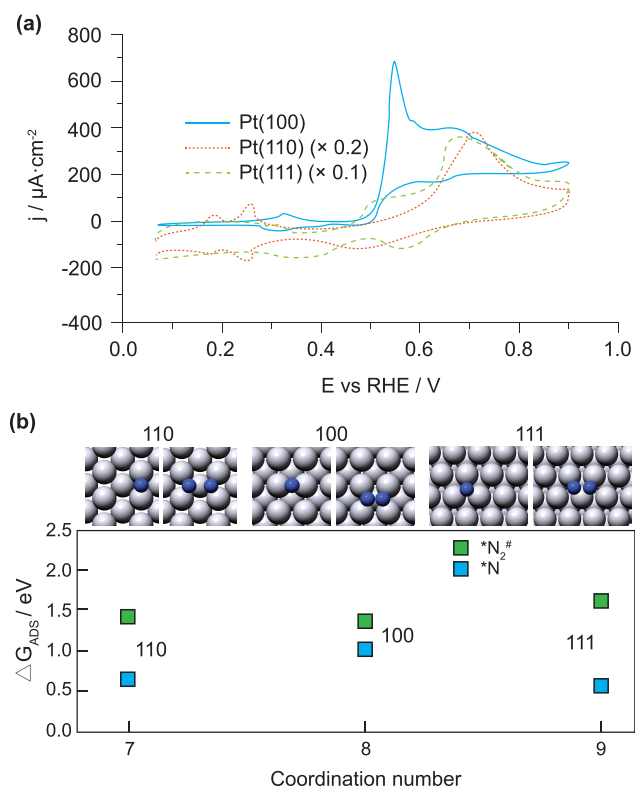


Figure 4. Structure sensitivity of AOR on Pt catalysts. (a) Voltammetric profiles of Pt(100), Pt(111), and Pt(110) single-crystal surfaces.¹⁹ (b) Adsorption energies of *N (blue) and the $\text{*N}_2^\#$ transition state (green) as a function of the regular coordination number (CN) of the active sites on different Pt surfaces.³⁹ Panel (a) is reproduced from ref 19 with permission. Copyright Elsevier. Panel (b) is reproduced from ref 39 with permission. Copyright American Chemical Society.

surface sites are less active in terms of activating NH_3 than (100) to begin with due to site coordination. Compared to Pt(111), *NH_2 on Pt(100) is more stable by 0.73 eV. However, a more important factor governing the structure sensitivity is its capability of binding *N , as shown in Figure 4b. On (111) and (110) sites, N binds strongly at the hollow and bridge sites, respectively, which prevents its removal through dimerization; for example, there is a 2.24 eV barrier for N–N coupling on Pt(111).¹⁷ Once *N is transiently generated, it blocks the sites for the further adsorption of NH_3 , resulting in diminishing current densities. However, on (100)-type sites, *N slightly prefers to adsorb on the bridge sites, at which *N dimerization across the hollow is facile (Figure 4b).^{9,10,17} The nearly degenerate adsorption of the bridge and hollow *N species on (100)-type sites eliminates the energy sink of the hollow sites found in the (111)-type counterparts, representing one key factor of the structure sensitivity of AOR on metal surfaces.

Role of Adsorbed Hydroxyl Species. The role of surface-adsorbed hydroxyl (OH) species in AOR kinetics, in comparison to that of bulk solution-phase OH^- , remains largely elusive. On Pt, *OH is considered as a relevant species because of its favorable formation starting at ~ 0.3 V vs RHE.^{40,41} At high potentials > 0.7 V vs RHE, *OH can serve as a passivation species on Pt electrodes (Figure 5a), thus inhibiting NH_3 oxidation.⁴¹ As a coadsorbed spectator species (Figure 5b), *OH has hydrogen bonding interactions with

*NH_2 and *NH , facilitating the dehydrogenation of *NH_2 by OH^- . The consideration of these subtle interactions drastically improves the agreement of theoretical calculations with experimental measurements in terms of the onset potential of AOR on Pt(100).⁹ As a proton acceptor (Figure 5c), the dehydrogenation reactivity of *OH and OH^- has been directly compared using ab initio molecular dynamics (AIMD) with slow-growth advanced sampling.⁴² Two different models were used to represent high potential (HP) and low potential (LP) systems controlled by the amount of potassium (K) atoms in the bulk solution. The effective electrode potential was calculated using the equation $U_{\text{SHE}} = (\Phi - \Phi_{\text{SHE}})/e$, where Φ is the ensemble-averaged work function of the system, and Φ_{SHE} (4.43 eV) is the work function of the standard hydrogen electrode.⁴³ The model consisted of bulk water molecules and OH^- ions above the surface to simulate the solid–liquid interface. It was determined that the adsorbed hydroxyl (*OH) was the reactive species during NH_3 dehydrogenation under the reaction conditions rather than the OH^- in bulk water.⁴² However, the electrode potential from AIMD simulations with one or two added K atoms is still much higher than the experimental potential range, and the energetics along the reaction coordinate of the dehydrogenation pathways can be compromised by possible side reactions involving explicit water molecules at the interface. As a buffer species (Figure 5d), it has been shown that the electrocatalytic activity of Pt catalysts can be significantly improved by modification with rare earth oxides, e.g., CeO_2 .⁴⁴ Additives with surface hydroxyl likely improve the supply capacity of *OH at the active Pt sites to participate in dehydrogenation or modulate reaction energetics. These results indicate that engineering active sites with OH-rich metal oxide moieties in their proximity can be one of the promising ways to design high-performance anode materials for AOR.

Surface Deactivation. Surface deactivation is a major challenge in electrocatalytic NH_3 oxidation on Pt-based catalysts. Computational modeling has provided deep insights into the underlying origin of deactivation processes. Initially, surface-adsorbed *N was considered as the poisoning species, as postulated in the G–M mechanism.³⁰ This is only plausible at (111)-type sites, in which their dimerization at hollow sites is kinetically prohibited.¹⁶ Intriguingly, at high operating potentials (> 0.63 V vs RHE), the dehydrogenation of *NH to *N becomes thermodynamically feasible on (100)-type surface sites,^{9,15,17} which coincides with the onset potential of surface deactivation. On Pt(100), however, *N binds rather weakly on the bridge sites, and their dimerization is barrierless,^{9,10} which suggests that *N is not a true poisoning species. The current hypothesis is that *N is only a precursor to the surface deactivation of Pt(100). At high potentials, surface sites have a high coverage of *OH , which easily couples with *N to form *NOH , the dehydrogenation of which leads to strongly bonded *NO .^{9,10} This is supported by differential electrochemical mass spectroscopy (DEMS)¹⁹ measurements, detecting both desorbed NO and N_2O on Pt(100) but not on Pt(110) and Pt(111) at potentials > 0.6 V (Figure 6a). In situ attenuated total reflection infrared (ATR-IR) spectroscopy (Figure 6b) suggests the formation of bridged NO on Pt electrodes above the onset potential of NH_3 oxidation, resulting in reaction inhibition and surface deactivation.³¹ To alleviate the poisoning issue, it is highly desirable to avoid the formation of *N because its coupling to surface *OH to form strongly bonded *NO seems inevitable. Weakening *NO

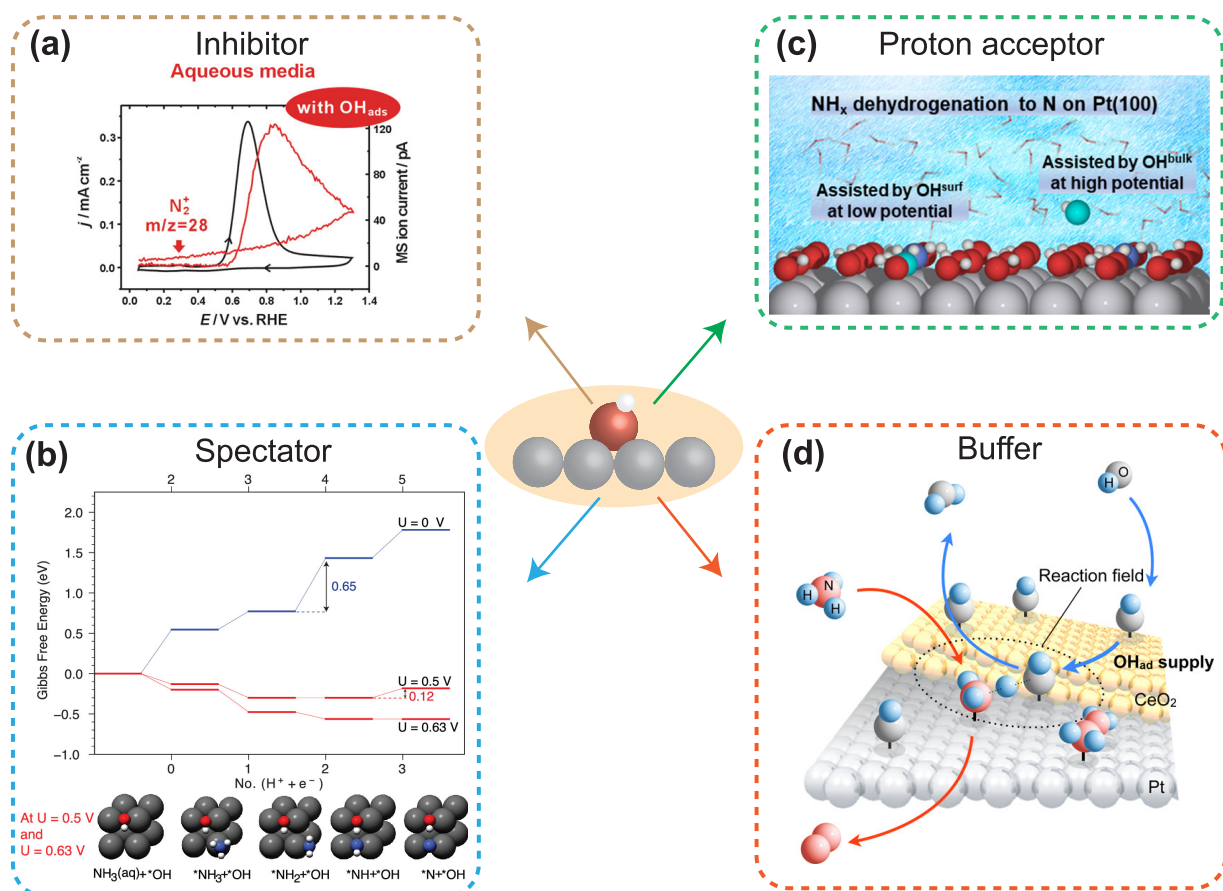


Figure 5. Role of surface-adsorbed hydroxyl species as (a) an inhibitor,⁴¹ (b) a spectator,⁹ (c) a proton acceptor,⁴² and (b) a buffer⁴⁴ in electrocatalytic NH_3 oxidation. Panel (a) is reproduced from ref 41 with permission. Copyright American Chemical Society. Panel (b) is reproduced from ref 9 with permission. Copyright American Chemical Society. Panel (c) is reproduced from ref 42 with permission. Copyright American Chemical Society. Panel (d) is reproduced from ref 44 with permission. Copyright American Chemical Society.

adsorption might also be a good strategy that allows its facile reduction if transiently formed. To mitigate surface deactivation, various strategies have been proposed, including the use of alloyed catalysts, surface modification, and co-catalysts.⁴⁵ To further theoretically screen catalytic materials, $\ast\text{N}$ and $\ast\text{NO}$ adsorption energies can be employed as two reactivity descriptors in a microkinetic model that includes the formation of precursor species in surface deactivation pathways, as shown in (Figure 3).

CURRENT LIMITATIONS AND CHALLENGES IN COMPUTATIONAL MODELING

Computational modeling has become an essential tool in many scientific fields including electrochemistry. However, there are limitations and challenges that must be addressed to improve the accuracy and reliability of computational models. In the current computational science paradigm, there is a lack of efficient ways to incorporate coverage effects of surface adsorbates into kinetics. Oftentimes, the coverage of reaction intermediates is assumed or only included on an ad hoc basis in microkinetic modeling. Including surface adsorbates self-consistently in microkinetics is a key step for improving ab initio kinetic models. Expectedly, it is extremely challenging to consider all possible intermediates at different coverages. Leveraging deep learning algorithms might hold the key to a new paradigm of catalysis modeling enabled by data

science.^{46–48} In terms of machine learning (ML), interpretable deep learning is particularly promising⁴⁹ because we can gather insights on electronic structure,^{50,51} surface chemisorption,⁵² and adsorbate–adsorbate interactions⁵³ from the rich physical knowledge of surface chemistry and catalysis. In a recent work by Pillai et al.,⁵⁴ the electronic structure properties that govern the surface reactivity of Pt-based AOR catalysts were unraveled by interpretable deep learning within the theory infused neural network (TinNet) framework.⁵⁵ In this study, the most important governing factor of nitrogen stabilization on the two Pt-based alloys Pt_3Ru and $\text{Pt}_{3/2}\text{Co}_{1/2}$ relative to Pt was found to be the adsorbate resonance energies of the frontier N_{2p} orbitals, followed by the conventional d-band center of the active site atoms. Both factors fine-tune the adsorption energies of nitrogen through the position and occupation of adsorbate–metal antibonding states formed by the hybridization of the surface d-state and renormalized adsorbate states.

The computational cost of atomistic simulations can be prohibitively expensive for large systems or long time scales. For example, ab initio atomistic modeling of the electrode–electrolyte interface is currently limited by the enormous computing it would take to describe the electric double layer accurately while also considering ion effects, electrode potential, pH, and solvation. To model rare events like chemical reactions, advanced sampling methods, such as replica-exchange molecular dynamics,⁵⁶ metadynamics,⁵⁷ and

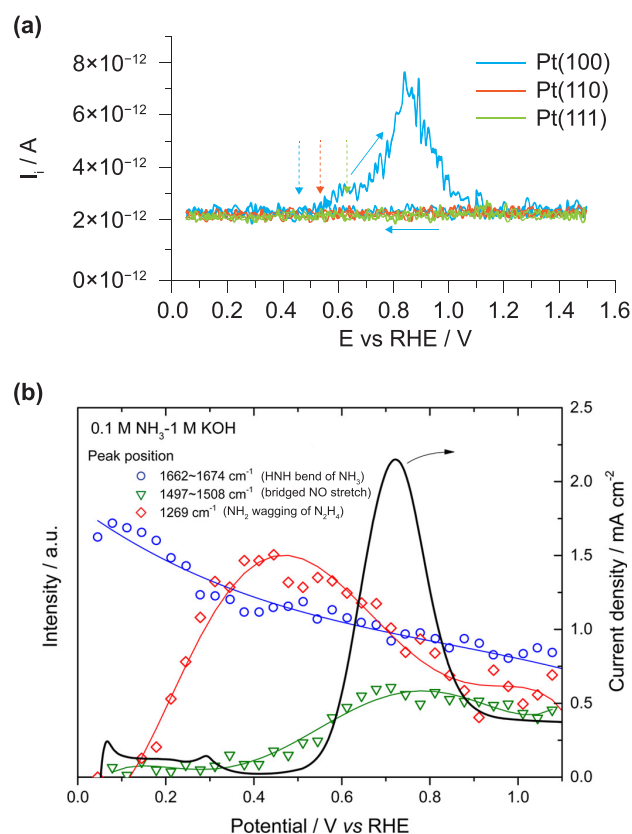


Figure 6. Surface deactivation of electrochemical NH_3 oxidation on Pt catalysts. (a) Mass spectrometric cyclic voltammograms (MSCVs) ($m/z = 31$, ^{15}NO) of Pt(100), Pt(110), and Pt(111) electrodes in 0.1 M NaOH and 10^{-3} M NH_3 after cycling up to 0.90 V.¹⁹ (b) In situ IR measurements of Pt thin films under NH_3 oxidation conditions.³¹ Panel (a) is reproduced from ref 19 with permission. Copyright Elsevier. Panel (b) is reproduced from ref 31 with permission. Copyright American Chemical Society.

blue moon ensemble,⁵⁸ are required to capture the dynamics of catalytic processes within an acceptable simulation time frame. One requirement for the accuracy and efficiency of such enhanced sampling methods is an accurate potential energy surface, which can potentially be provided by well-trained machine learning potentials.^{59,60} The development of highly accurate and efficient machine learning potentials by learning from ab initio data for describing electrocatalytic processes is promising.⁶⁰ Improvements to algorithms^{61,62} and computational data ecosystems^{63,64} will help to discover more about the phenomena buried at the solid–electrolyte interfaces.

Finally, the complexity of electrochemical systems, such as the presence of multiple reaction pathways and the coupling of physical processes at different scales, e.g., mass transport, can also pose challenges for computational modeling. Addressing these limitations and challenges will require interdisciplinary collaborations between experimentalists and theorists as well as the development of new theoretical and computational methods.^{48,65}

FUTURE OUTLOOK AND OPPORTUNITIES

The field of electrochemical NH_3 oxidation (AOR) has seen significant advances in recent years, largely attributed to the widespread adoption of computational modeling. As computational power and algorithms continue to improve, there is great

potential for further insights into AOR and the development of more efficient catalysts. For example, the use of hybrid functionals in DFT calculations can improve the accuracy of predictions, while the development of more efficient algorithms that reduce computational costs is required for enabling larger-scale simulations. One emerging technique is to use ML algorithms that can predict material properties by learning from data. By training ML models on large data sets of known catalysts and their properties, ML algorithms can transfer the learned knowledge by fine-tuning with a small but more accurate data set in a transfer learning scheme,^{66,67} making accurate predictions about the properties of new materials. Compared to a first-principles only approach, computational costs can be significantly lowered by ML algorithms once they are trained with data. Machine learning has already been used successfully to find improved AOR catalysts¹¹ and for other advances of catalysis science.⁴⁶ Another promising technique is data-enhanced multiscale modeling,⁴⁸ which combines different levels of modeling techniques and data science to provide a more comprehensive understanding of catalytic processes. For example, molecular dynamics simulations can be used to study the behavior of a collection of atoms and molecules, while continuum models can provide insights into the behavior of the systems by considering transport processes. A data-enhanced multiscale modeling technique can be used to develop a better understanding of the solid–electrolyte interface with thorough, yet efficient, computational approaches. Probing the multiscale behavior of the solid–electrolyte interface will deepen our fundamental understanding of AOR (or charge transfer reactions in general) and may lead to its optimization toward practical applications.

The future of computational modeling is bright with many opportunities for further insights. By continuing to develop and refine computational techniques, the discovery timeline of new catalytic materials with improved AOR efficiencies can be greatly shortened, meeting the need for global efforts toward decarbonization. Resolving key steps in surface deactivation and finding strategies for significantly improving surface stability are vital for the future implementation of this technology in energy and environmental applications.

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Notes

The authors declare no competing financial interest.

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