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Environmental Chemistry PERSPECTIVE

The Innovation Materials

Can environmental catalysts achieve both high activity and stability simultaneously?

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Advanced oxidation processes (AOPs) efficiently degrade water pollutants, but their catalysts often prioritize activity over long-term stability, posing challenges for sustained wastewater treatment. High activity and stability are thermodynamically conflicting, with active catalysts degrading quickly and raising costs. This paper explores technologies optimizing both aspects, aiming for sustainable, cost-effective wastewater solutions.

INTRODUCTION

While industrial production fulfills fundamental material demands, it also brings serious environmental issues, notably through the release of industrial by-products into natural ecosystems. The water pollution and resource scarcity pose major threats to global sustainability, highlighting the critical importance of treating organic wastewater. Among AOPs, the Fenton and Fenton-like reactions have made significant progress in both basic research and application for organic wastewater treatment. The Fenton reaction, discovered by H.J. Fenton in 1894, demonstrated that ferrous salts could activate hydrogen peroxide (H_2O_2) to produce hydroxyl radicals (·OH) for pollutant degradation. Fenton-like reactions extend this by using metal ions like Cu^{2+}, Co^{2+}, and Mn^{2+}, to activate H_2O_2, peroxymonosulfate (PMS) and peroxydisulfate (PDS), producing reactive oxygen species (ROSs) for enhanced pollutant degradation. Recently, heterogeneous systems have garnered more attention than homogeneous systems, due to their lower metal leaching rates and greater stability with performance mainly dependent on the catalyst and oxidant properties.

Balancing high activity and long-term stability in the degradation system is challenging, particularly in real-world applications. High catalyst stability enhances oxidant utilization, reduces chemical dosage, and lower operating costs, while high activity ensures efficient treatment and system performance. High activity typically requires high surface energy or active redox, which, while enhancing reactivity, can lead to material degradation or failure—especially in complex environments with interfering by-products. Thermodynamically, high activity and stability often conflict, as highly active catalysts with high surface energy are more vulnerable to environmental factors, leading to structural and electronic disruptions that reduce activity. Recent research has focused on designing catalysts that address this challenge by modulating active sites—adjusting charge properties, adsorption energies, and reaction pathways—thereby optimizing activation and free energies for long-term use.

Although catalyst activity and stability are thermodynamically contradictory, it is feasible to develop catalytic systems that reside in both high activity and longevity. Current research focuses on balancing activity and stability through mechanisms like intermolecular electron transfer in organic pollutants, co-catalytic Fenton systems, and amphoteric metals to enhance stability. These strategies aim to optimize performance and longevity for more effective and sustainable wastewater treatment.

Intermolecular electron transfer in pollutants, catalysts and oxidants

Organic pollutants in water can be degraded through many mechanisms, among which the intermolecular electron transfer mechanism is considered the most promising. In this mechanism (Figure 1A) , the electrons from the organic pollutant transfer to the catalyst, which acts as a bridge to deliver the electrons to the active site, thereby promoting degradation and reducing the consumption of oxidants for long-lasting performance.

For example, Wang et al. (2023)[1] used graphene oxide (GO) a to enhance Mn(VII)-mediated pollutant removal, where Mn(VII) adsorbs onto GO's hydroxyl groups, boosting redox potential. GO mediates electron transfer as sulfisoxazole (SSX) adsorbs via π-π interactions, donating electrons to GO, while Mn(VII) accepts them. The system effectively degrades bisphenol A (BPA), SSX, aniline and carbamazepine (CBZ) showing complete SSX removal over 10 cycles, with Mn(VII) replenishment. Future research should focus on developing carbon catalysts with larger particle sizes and tailored functional groups. Moreover, Chen et al. (2023)[2] developed a single-atom catalyst ($SAMCC_{0.5}$) for efficient PMS activation and phenol degradation. Phenol donates electrons to Mo sites, which transfer them to Co sites via a conjugated framework, increasing electron density without Co losing electrons. This enables cyclic phenol degradation, enhancing both activity and stability. The $SAMCC_{0.5}$/PMS system remains effective under extreme pH conditions for up to 10 days, efficiently degrading high concentrations of phenolic pollutants.

Construction of catalytic Fenton systems

Fenton technology has advanced in wastewater treatment but still faces challenges, such as low Fe^{2+}/Fe^{3+} recycling efficiency and iron sludge formation causing secondary pollution. Iron-based catalytic systems generate ROSs via Fe(III)/Fe(II) cycling, with Fe(II) recovery being the rate-determining step. Adding co-catalysts accelerates iron cycling, inhibits iron sludge formation and enhances system activity and long-term stability, achieving a balance between catalytic activity and stability.

Rechargeable Fenton system

Polyaniline (PANi), a redox-active polymer, exhibits reversible electron storage and release capabilities during charge-discharge cycles. This behavior mainly stems from active sites like conjugated amines, nitroxide radicals, and conjugated carbonyls. Zhou et al. (2023)[3] designed a chargeable Fenton reaction system where electron-rich PANi is continuously discharged during the Fenton reaction, transferring electrons to Fe^{3+} and generating abundant ·OH radicals for organic pollutant degradation. Simultaneously, electron carriers in the form of H atoms are released into solution, causing a decrease in pH (Figure 1B). PANi's planar structures increases molecular conjugation during the reaction, stabilizing the oxidation products and enhancing its electron-donating capacity. Consequently, PANi with high stability in both oxidized and reduced states and low energy barriers outperforms other redox-active polymers in reducing Fe^{3+}. Inspired by these studies, the redox-active polymers can be introduced as electron shuttles to accelerate the electron transport, balancing system activity and stability.

MoS_2-catalyzed Fenton system

MoS_2, as a co-catalyst, can promote iron cycling in Fenton systems. Its graphene-like structure provides high catalytic activity, excellent stability, and low cost, making it an ideal candidate for Fenton-like reactions. Additionally, MoS_2 enhances oxidative degradation when integrated with oxidants such as H_2O_2, PMS, and PDS, further demonstrating its potential.

For instance, Yan et al.(2021)[4] developed a MoS_2 co-catalytic heterogeneous Fenton system ($CoFe_2O_4/MoS_2$). In this system, the Mo^{4+} exposed on the surface of MoS_2 enhances Fe^{3+}/Fe^{2+} cycling at the solid-liquid interface and prevents catalyst deactivation, while the oxidant H_2O_2 facilitates the Mo^{6+}/Mo^{4+} redox cycle. The sulfur in MoS_2 creates an acidic microenviron-

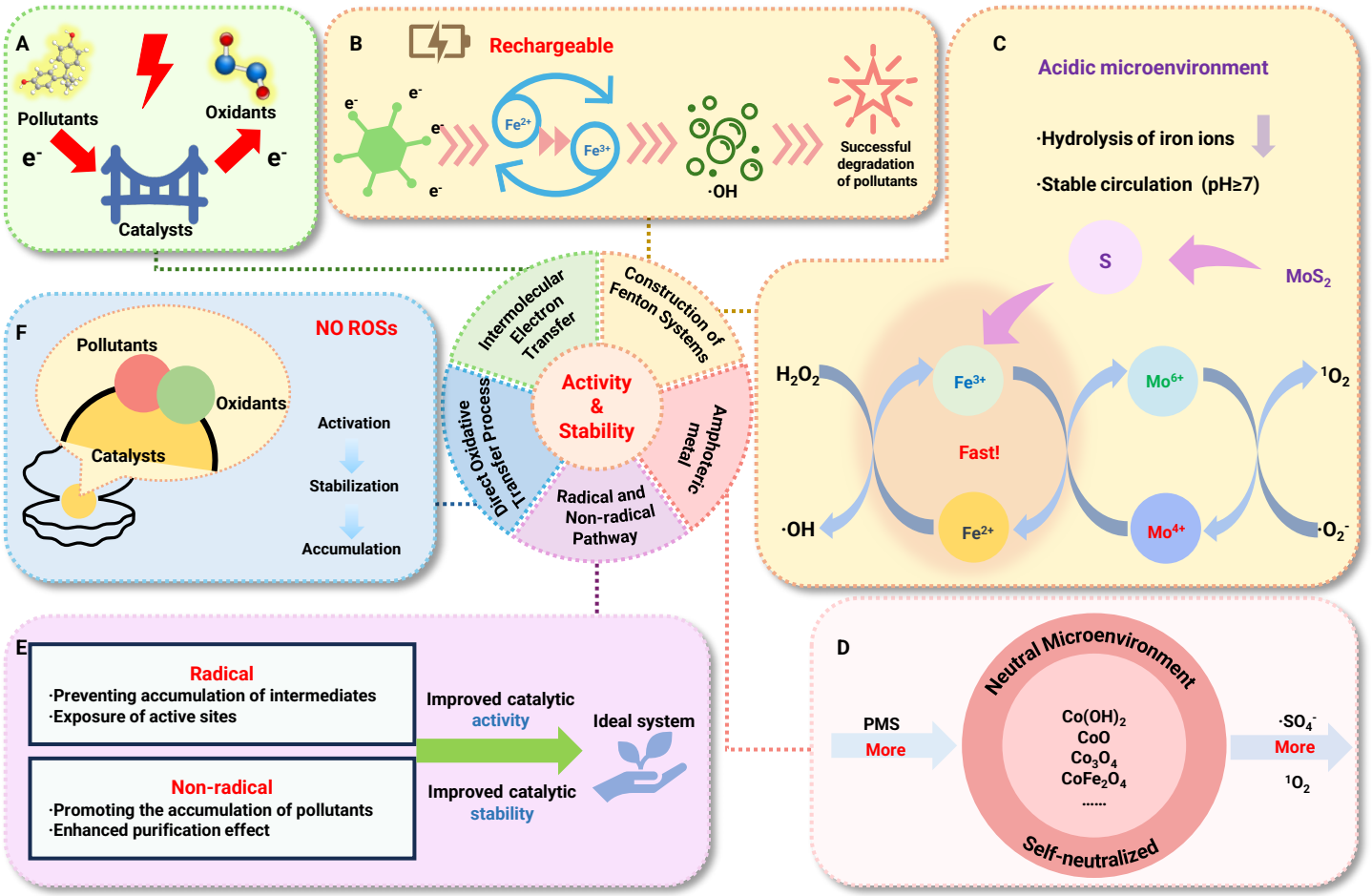

Figure 1. Balancing reactivity and stability in oxidative degradation systems (A) Intermolecular electron transfer mechanism; (B) Rechargeable Fenton system; (C) MoS_2-catalyzed Fenton system; (D) Amphoteric metal; (E) Combined radical and non-radical pathway; (F) Mechanism of DOTP.

ment on the catalyst's surface, inhibiting iron ion hydrolysis and enhancing stability (Figure 1C). This microenvironment remains stable regardless of surrounding pH, ensuring consistent Fe^{3+}/Fe^{2+} cycling even under neutral or alkaline conditions.

While MoS_2 improves iron cycling and reduces sludge in Fenton reactions, its powdered form hinders recycling. Additionally, MoS_2 may react with H_2O_2, releasing metal ions and causing heavy metal pollution. Future research could focus on immobilizing MoS_2 on suitable carriers to enhance reusability or exploring alternative co-catalysts for improved system longevity.

Boron-promoted Fenton system

Inorganic co-catalysts are crucial in providing green electron donors to facilitate iron recycling, for efficient iron ions conversion. The Fe(III)-based system is better stabilized but less active than the Fe(II)-based ones. Zhou et al. (2020)[5] proposed a boron-enhanced Fenton system, where B-B and low-oxide boron donate electrons to Fe(III), generating Fe(II), while inert boron oxide slows Fe(II) fomation. This system reduces iron salt usage, enhances H_2O_2 utilization, and outperforms other reducing agent-assisted Fenton systems in oxidation capacity.

Although this system effectively enhances the iron cycle, we cannot ignore the potential toxicity of leaching boron. Future studies should include toxicity and economic analyses and develop water treatment processes to mitigate boron-related health and environmental risks. Since boron concentration limits in drinking water vary globally, further research is needed for different application scenarios. The World Health Organization (WHO) has a boron concentration limit of 2.4 mg/L, while Chinese《Standards for drinking water quality》(GB 5749-2022) has a boron concentration limit of 0.5 mg/L.

Amphoteric metal properties

Environmental pH significantly influences PMS activity, and maintaining a stable, moderate pH enhances reaction efficiency. Bao et al. (2023)[6] synthesized zinc cobalt basic carbonate to create a neutral microenvironment at the cobalt-based catalyst interface, improving PMS decomposition without altering the main active species (Figure 1D). The Zn-CHH component enabled self-regulation across a wide pH range (3-11), facilitating efficient phenol degradation. This catalyst shows good stability, as its activity could be restored in long-term experiments by adding an alkali. Moreover, common anions and varying water qualities had minimal impact, demonstrating its potential for the effective deep treatment of real nitrobenzene wastewater. Similarly, Zhang et al. (2024)[7] prepared spinel $ZnCo_2O_4$ porous nanosheets with different Co/Zn ratios by hydrothermal calcination. The Zn-rich structures, combined with the amphoteric $\equiv Zn(OH)_2$ to create a neutral microenvironment, broadening the applicable pH for PMS activation. In alkaline environments, Zn^{2+} is converted to $\equiv Zn(OH)_2$. In neutral or acidic environments, the Zn-rich structures undergo $\equiv ZnO_2$ hydrolysis to $\equiv Zn(OH)_2$, followed by proton neutralization.

Combined radical and non-radical pathway

Radicals prevent intermediate accumulation, keeping active sites exposed for continuous catalysis, while non-radical reactions promote surface-limited purification. Their synergy outperforms either pathway alone by maximizing oxidation capacity and adaptability. This balance enhances activity and stability, ensuring long-term catalytic performance (Figure 1E).

Guo et al.(2023)[8] constructed a heterogeneous Fenton-like system, A/C-$CoMnO_x$, optimizing structure via crystallization engineering while maintaining morphology and composition. Its high performance stems from A and C domain synergy at three levels: (1) Molecular adsorption: The A/C interface enhances BPA binding at Co_{Oh} sites and PMS adsorption at Mn_{Oh} sites, improving PMS utilization. (2) Electronic interaction: Abundant Mn^{3+}/Mn^{4+} species facilitate electron transfer, boosting redox cycling of Mn and Co. (3)

Reactive species: Mixed pathways address surface passivation, where radicals degrade surface-bound pollutants, and ETP enhances pollutant enrichment for effective degradation.

Catalyst surface passivation can reduce activity over time, but a hybrid reactive species approach mitigates this issue. Radicals degrade pollutants and prevent intermediate accumulation, while ETP enriches pollutant for effective degradation. Synergistic interactions at the A/C interface and between radicals and ETP ensure the activity and stability of the A/C-$CoMnO_x$/PMS system, ensuring perdormance in treating tap water, lake water, and electroplating wastewater.

Direct oxidative transfer process (DOTP)

Zhang et al. (2022)[9] demonstrated that the DOTP achieves high catalytic performance and stability through its unique oxidation mechanism (Figure 1F). It effectively removes pollutants with electron-donating groups, requiring only low doses of oxidants and no external energy. Unlike adsorption, DOTP works without high-surface-area nanomaterials, enabling the use of cost-effective nanocatalysts. Building on these findings, the researchers proposed a metal-oxygen halide (MOX)-based Fenton system[10] for the coupling and polymerization of organic pollutants through mild surface oxidation, avoiding the generation of reactive oxygen species and reducing the consumption of H_2O_2 while minimizing the generation of toxic by-products. This system leverages O-bridged M and X sites to selectively activate contaminants and H_2O_2. Combined with low-cost MOX materials like BiOI, FeOCl, and VOCl, it integrates with dynamic membrane filtration for energy-efficient removal of micropollutants. Its affordability and efficiency make it ideal for resource-limited and by-product-sensitive water treatment, complementing traditional AOPs.

PERSPECTIVE

This paper highlights key advancements in water pollutant degradation, providing valuable insights for the future of catalyst development:

Prioritizing activity, durability, and cost-effectiveness: Future catalyst development will aim to balance high activity and long-term stability through optimized designs, appropriate support materials, and the incorporation of co-catalysts, amphoteric metals, or novel mechanisms like DOTP. These strategies aim to enhance efficiency, durability, and cost-effectiveness for practical applications.

Extending longevity for industrial applications: Developing durable, high-performance catalysts is crucial for industrial applications. Current longevity tests (5 to 10 days) are insufficient for real-world evaluation. Future research should extend tests to 15 days or longer to better simulate industrial conditions and assess long-term stability and performance.

Enhancing Fenton system stability: While individual Fenton catalysts may exhibit instability, the overall Fenton system can gain stability through co-catalysts and oxidants. This approach effectively enhance both activity and stability of advanced oxidation technologies (AOTs).

Addressing the complexity of real wastewater: In sewage treatment, catalyst evaluation should account for real wastewater complexity. Experiments should be conducted using actual samples rather than simplified, single-component models. Tailored technologies focusing on activity, stability, and selectivity, are needed for diverse pollutants in real-world scenarios.

Flexibility in catalytic oxidation systems: Catalytic oxidation systems offer flexibility in water treatment. Their optimization and catalysts design should be targeted to the specific needs of each application.

Future research should focus on developing catalysts that balance high activity and stability, overcoming the "see-saw" effect. This will facilitate more novel systems, achieving higher activity, greater stability, and broader environmental applicability.

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AUTHOR CONTRIBUTIONS

All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.