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Coordination Chemistry of Cobalt in Peroxymonosulfate Activation for Pharmaceutical Contaminant Removal from Wastewater: Linking Active-Site Structure, Activation Mechanisms, Environmental Stability, and Practical Applicability—A Critical Review

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ABSTRACT
Pharmaceutical residues in wastewater represent an important class of contaminants of emerging concern because of their continuous discharge into aquatic environments, incomplete removal by conventional wastewater-treatment processes, biological activity at low concentrations, and potential transformation into products of uncertain environmental significance. These concerns have stimulated increasing interest in advanced oxidation processes (AOPs), particularly systems based on peroxymonosulfate (PMS) activation, for the treatment of pharmaceutical micropollutants.
This critical review examines cobalt-mediated PMS activation from a coordination-chemistry perspective and evaluates how the local structure and electronic properties of cobalt active sites influence PMS activation mechanisms, pharmaceutical degradation, catalyst stability, cobalt leaching, and practical wastewater-treatment performance. Particular emphasis is placed on establishing mechanistically defensible relationships among cobalt coordination environment, electronic structure, PMS adsorption and activation, dominant oxidation pathways, and treatment outcomes.
The classical interpretation of Co/PMS chemistry emphasizes Co(II)/Co(III) redox cycling and the formation of sulfate and hydroxyl radicals. Contemporary evidence, however, demonstrates substantially greater mechanistic diversity, including reactive Co-PMS complexes, surface-mediated processes, interfacial electron transfer, nonradical pathways, and proposed high-valent cobalt intermediates. The relative importance of these pathways depends on cobalt speciation, coordination environment, catalyst support, reaction conditions, substrate properties, and water-matrix composition. Because several proposed intermediates remain difficult to identify unambiguously, mechanistic assignments require converging evidence rather than reliance on radical-scavenging experiments alone.
Recent coordination-engineering studies further demonstrate that the identity, coordination number, and spatial arrangement of atoms surrounding cobalt can modify its electronic structure, PMS-binding configuration, electron-transfer behavior, peroxide-bond activation, and dominant oxidation pathway. Coordination engineering therefore offers a rational strategy for controlling not only catalytic reactivity and selectivity but also cobalt stabilization.
Beyond intrinsic catalytic activity, this review critically evaluates the effects of pH, catalyst loading, PMS dosage, pollutant properties, inorganic ions, natural and effluent organic matter, temperature, and water-matrix complexity. Particular attention is given to PMS utilization, mineralization, transformation products, post-treatment toxicity, cobalt leaching, catalyst recovery and reuse, and the transition from model aqueous systems to real wastewater.
Active-site structure → PMS activation mechanism → chemical treatment performance → catalyst stability → environmental safety → practical applicability
The analysis indicates that future Co/PMS research should move beyond maximizing parent-pharmaceutical disappearance and instead focus on engineering cobalt coordination environments that simultaneously promote efficient PMS utilization, meaningful contaminant degradation and mineralization, low cobalt release, structural stability, environmental safety, and reliable performance under realistic wastewater conditions.
Keywords: pharmaceutical contaminants; wastewater treatment; cobalt; peroxymonosulfate; coordination chemistry; advanced oxidation processes; active-site engineering; nonradical oxidation; cobalt leaching; catalyst stability.

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I. Introduction

The extensive use of pharmaceuticals in human and veterinary medicine has resulted in their continuous introduction into municipal, hospital, agricultural, and industrial wastewater streams. Parent pharmaceuticals and their metabolites may enter wastewater through human and animal excretion, improper disposal of unused medicines, healthcare discharges, veterinary activities, and pharmaceutical-manufacturing effluents [1,2].

A broad range of pharmaceuticals has consequently been detected in wastewater and receiving aquatic environments, including antibiotics, analgesics, anti-inflammatory drugs, hormones, antidepressants, beta-blockers, and other biologically active compounds. Although many occur at trace concentrations, their repeated introduction, biological activity, persistence or pseudo-persistence, and transformation behavior make them relevant to water-quality management and environmental-risk assessment [1].

Conventional wastewater-treatment plants are primarily designed to remove bulk organic matter, suspended solids, nutrients, and pathogens rather than trace organic micropollutants. Pharmaceutical removal therefore varies considerably according to molecular structure, physicochemical properties, biodegradability, sorption behavior, treatment configuration, microbial-community composition, and operating conditions [1–3].

The issue is directly relevant to Kuwait. Gevao et al. [4] investigated antibiotics in influent and effluent streams from wastewater-treatment plants in Kuwait and reported the occurrence of several compounds, including sulfamethoxazole, ciprofloxacin, clarithromycin, and cephalexin. The persistence of selected pharmaceuticals following treatment provides a locally relevant rationale for investigating advanced treatment processes capable of addressing pharmaceutical micropollutants.

Advanced oxidation processes have attracted considerable interest because they can chemically transform organic contaminants rather than merely transfer them between phases [2,3]. Among these technologies, PMS-based oxidation has emerged as an important research area. PMS can be activated through thermal, photochemical, electrochemical, and catalytic routes to generate reactive pathways capable of oxidizing a broad range of organic contaminants.

Cobalt is a highly reactive transition-metal activator of PMS [5]. Although the conventional description emphasizes Co(II)/Co(III) redox cycling and sulfate-radical formation, later work has shown that this picture is not universally sufficient. Depending on cobalt speciation and the local chemical environment, reactive Co-PMS complexes, surface-associated species, interfacial electron-transfer processes, nonradical routes, and cobalt intermediates in different oxidation states may contribute to oxidation [6,9–11].

Advances in catalyst characterization and active-site engineering have also shifted attention from bulk cobalt content to the local chemistry of catalytically accessible cobalt centers. Coordination number, first-shell ligands, local geometry, oxidation state, electronic structure, and metal-support interactions can alter PMS binding and activation [8,15,16]. The same structural variables can influence cobalt retention, making coordination chemistry relevant to both reactivity and leaching control.

This relationship creates an important bridge between coordination chemistry and wastewater-treatment engineering. A catalyst that rapidly removes a pharmaceutical in ultrapure water cannot necessarily be considered suitable for practical treatment if it requires excessive PMS, releases cobalt into the treated water, produces persistent or toxic transformation products, loses activity during reuse, or performs poorly in complex wastewater.

Accordingly, this review critically evaluates cobalt-mediated PMS activation through an integrated structure-mechanism-performance-stability-safety-application framework.

1.1. Research Problem

Co/PMS performance is frequently evaluated using the percentage disappearance of a selected pharmaceutical. Although useful, this metric alone does not adequately represent overall treatment effectiveness.

Disappearance of the parent compound does not necessarily indicate substantial mineralization. Oxidation can generate transformation products whose persistence and toxicity differ from those of the original pharmaceutical. Likewise, high apparent catalytic activity may coexist with excessive PMS consumption, cobalt release, catalyst reconstruction, or progressive deactivation.

A second limitation is the widespread use of simplified model solutions. Real wastewater contains dissolved organic matter, inorganic ions, nutrients, suspended or colloidal constituents, and mixtures of organic contaminants that can consume oxidant, quench reactive species, alter catalyst surfaces, or modify cobalt speciation.

Mechanistic interpretation presents an additional challenge. Although radical pathways remain important, increasing evidence supports the participation of Co-PMS complexes, surface-bound species, electron-transfer pathways, and other nonradical processes. The identity and relative importance of proposed high-valent cobalt intermediates also require careful experimental discrimination [9–11].

How does the coordination environment of cobalt govern PMS activation, and can this molecular-level control be translated into efficient, stable, environmentally responsible, and practically relevant treatment of pharmaceutical contaminants in wastewater?

1.2. Analytical Hypothesis

This review is guided by the hypothesis that modification of the local coordination environment of cobalt changes the electronic structure of the active site and its interaction with PMS, thereby influencing activation pathways, oxidant utilization, catalytic selectivity, structural stability, and cobalt leaching. This hypothesis is used as an analytical framework rather than being assumed to represent a universally established relationship across all cobalt-based PMS systems.

1.3. Objectives

The primary objective of this review is to critically examine the relationship between cobalt coordination chemistry and PMS activation and to determine how this relationship influences pharmaceutical-contaminant treatment.

1. Critically evaluate radical, nonradical, complex-mediated, and proposed high-valent pathways involved in cobalt-mediated PMS activation.
2. Examine relationships among cobalt coordination environment, electronic structure, PMS interaction, and catalytic behavior.
3. Assess the influence of operating conditions and water-matrix composition on Co/PMS performance.
4. Compare homogeneous, heterogeneous, supported, and coordination-engineered cobalt systems from a wastewater-treatment perspective.
5. Evaluate relationships among active-site engineering, catalyst stability, cobalt leaching, and reusability.
6. Examine mineralization, transformation products, and post-treatment toxicity as indicators of treatment effectiveness.
7. Assess the transition from simplified model systems to real wastewater.
8. Identify research priorities for environmentally safer and practically applicable Co/PMS technologies.

1.4. Scientific Contribution and Scope

Cobalt-mediated PMS activation, radical and nonradical pathways, cobalt leaching, and coordination engineering have individually received substantial attention [6,12,13,16]. The contribution of this review therefore does not rest on claiming novelty for these concepts themselves.

Its contribution is instead the critical integration of coordination-level evidence with wastewater-treatment engineering and environmental-safety endpoints through the following causal framework:

Cobalt coordination environment → electronic structure → PMS interaction and activation → oxidation pathway → pharmaceutical degradation and mineralization → catalyst stability and cobalt leaching → treated-water quality → practical applicability

The central evaluative question consequently shifts from "Which catalyst achieves the highest pollutant-removal efficiency?" to "How does the chemical structure of the cobalt active site control PMS activation, and does this mechanistic advantage translate into safer, more stable, and more efficient wastewater treatment?"

II. Methodology of the Critical Review

This article was designed as a critical review rather than a systematic review in the strict methodological sense. Its purpose is to synthesize and critically evaluate evidence linking cobalt coordination chemistry with PMS activation mechanisms, pharmaceutical degradation, catalyst stability, and wastewater-treatment applicability.

The literature considered spans four interconnected areas: pharmaceutical contaminants in wastewater; fundamental PMS activation chemistry; homogeneous and heterogeneous cobalt-based catalytic systems; and environmental stability and practical application of Co/PMS processes.

Relevant concepts included pharmaceutical contaminants, wastewater, cobalt catalysts, cobalt-based catalysts, peroxymonosulfate, PMS activation, cobalt coordination environment, sulfate-radical chemistry, nonradical oxidation, Co-PMS

complexes, high-valent cobalt, pharmaceutical degradation, cobalt leaching, catalyst stability, single-atom cobalt, and real wastewater.

Priority was given to peer-reviewed original research when mechanistic, structural, or quantitative performance claims were evaluated. Review articles were used primarily to establish the broader state of knowledge and identify areas of consensus or controversy, while seminal studies were retained where necessary to trace the development of mechanistic understanding.

Because published studies differ substantially in pollutant concentration, catalyst loading, PMS dosage, pH, reaction time, temperature, and water matrix, cross-study comparison based solely on removal percentage was avoided. Evidence was instead interpreted using multiple criteria, including catalyst architecture; cobalt coordination environment where experimentally supported; target contaminant; operating conditions; proposed activation pathway; strength of mechanistic evidence; degradation and mineralization; transformation products; toxicity; cobalt leaching; catalyst reuse; and matrix complexity.

Particular caution was applied to mechanistic assignments based exclusively on radical-scavenging experiments. Scavengers differ in reaction kinetics, selectivity, accessibility to surface-associated species, and possible effects on the reaction environment. Stronger mechanistic assignments therefore require converging evidence from complementary approaches such as electron paramagnetic resonance (EPR), selective chemical probes, kinetic analysis, isotope-based experiments, electrochemical measurements, X-ray absorption spectroscopy (XAS), in situ or operando characterization, controlled structural comparisons, and computational analysis where appropriate.

The evidence was ultimately interpreted according to the sequence: Active-site structure → PMS activation mechanism → treatment performance → catalyst stability → environmental safety → practical applicability. This approach distinguishes materials that merely exhibit high apparent activity from systems for which catalytic performance can be rationally connected to active-site chemistry and environmentally relevant treatment outcomes.

III. Pharmaceutical Residues in Wastewater and the Need for Advanced Treatment

3.1. Sources and Environmental Relevance

Pharmaceutical residues enter wastewater through domestic, medical, veterinary, and industrial pathways. Following administration, parent compounds and metabolites can be excreted and transported into municipal sewer systems. Additional inputs arise from improper disposal, healthcare facilities, veterinary activities, and pharmaceutical-manufacturing discharges [1,2]. Repeated pharmaceutical consumption can generate continuous environmental inputs; consequently, even compounds that are not intrinsically highly persistent may exhibit pseudo-persistent occurrence when their rate of introduction exceeds their rate of environmental removal. Their environmental relevance therefore depends not only on concentration but also on biological activity, exposure frequency, transformation behavior, and potential effects on aquatic organisms.

3.2. Limitations of Conventional Treatment

Conventional biological wastewater treatment can remove some pharmaceuticals through biodegradation, biotransformation, and sorption to sludge. Removal, however, is compound-specific and process-dependent [1–3]. Molecular structure, ionization, hydrophobicity, biodegradability, hydraulic and solids retention times, temperature, microbial-community composition, and operating conditions can all influence pharmaceutical fate. Effective removal of bulk organic matter and nutrients therefore does not guarantee effective elimination of trace pharmaceutical contaminants. This limitation provides an important rationale for advanced treatment processes targeting compounds that persist following conventional treatment.

3.3. Adsorption and Membrane Separation

Activated carbon and other porous adsorbents can substantially reduce aqueous concentrations of many pharmaceuticals. Similarly, nanofiltration and reverse osmosis can provide high rejection of numerous pharmaceutical contaminants. These processes are important treatment options, but they predominantly separate contaminants rather than necessarily destroy their molecular structures. Adsorption requires regeneration or appropriate management of spent media, whereas membrane processes generate concentrate streams that require further treatment or disposal. Thus, high removal from the treated-water stream should not automatically be interpreted as contaminant destruction.

3.4. Advanced Oxidation Processes

AOPs provide a complementary treatment strategy because their objective is to chemically transform organic contaminants. Technologies investigated for pharmaceutical removal include ozonation, UV/H2O2, Fenton and photo-Fenton processes, photocatalysis, and persulfate- or PMS-based oxidation [2,3,14]. PMS-based systems are particularly attractive because PMS can be activated

through several routes and can participate in both radical and nonradical oxidation chemistry. Nevertheless, chemical oxidation should not automatically be equated with complete environmental remediation. Three outcomes must be clearly distinguished: Parent-compound disappearance → molecular transformation/degradation → mineralization. This distinction is fundamental when evaluating cobalt-mediated PMS activation.

IV. Cobalt-Mediated Activation of Peroxymonosulfate

4.1. Fundamental Co/PMS Chemistry

The classical interpretation of cobalt-mediated PMS activation is based on electron transfer from Co(II) to PMS, accompanied by oxidation of Co(II) to Co(III) and generation of sulfate radicals [5]. A simplified representation is:

$$Co^{2+} + HSO_5^- \longrightarrow Co^{3+} + SO_4^{\bullet-} + OH^-$$

Scheme 1. Classical Co(II)-mediated activation of peroxymonosulfate (PMS).

Sulfate radicals can oxidize organic compounds through electron transfer, hydrogen abstraction, and addition reactions. Depending on the aqueous environment, secondary chemistry may also generate hydroxyl radicals. This classical redox model remains an important foundation for understanding Co/PMS chemistry. However, it should be regarded as a simplified representation rather than a universal mechanism. Contemporary evidence indicates that PMS activation can involve multiple parallel or competing pathways whose relative importance depends on cobalt speciation, coordination environment, catalyst structure, substrate characteristics, reaction conditions, and water-matrix composition [6,9–11].

4.2. Radical Activation Pathways

Sulfate and hydroxyl radicals remain important oxidants in many Co/PMS systems. Their relative contributions depend on cobalt speciation, catalyst structure, pH, PMS concentration, substrate identity, and background water chemistry. Radical pathways can provide rapid and broad-spectrum oxidation. Their high reactivity, however, can also promote nonselective consumption by natural or effluent organic matter and inorganic constituents. Consequently, high radical production in ultrapure water does not necessarily translate into efficient oxidant utilization in real wastewater. The relevant engineering parameter is therefore not radical generation alone but the fraction of oxidative capacity that contributes usefully to contaminant degradation and mineralization.

4.3. Nonradical and Surface-Mediated Pathways

Evidence for nonradical PMS activation has expanded considerably [6,8,9,15,16]. Proposed pathways include surface-bound activated species, interfacial electron transfer, reactive PMS complexes, singlet-oxygen-associated chemistry, and proposed high-valent metal species. Importantly, the term nonradical should not be treated as a single mechanistic category. Demonstrating that freely diffusing sulfate or hydroxyl radicals are not dominant does not identify the actual oxidizing intermediate. Some nonradical systems may exhibit greater selectivity or improved resistance to matrix interference, but these advantages are system-specific and require experimental verification. Nonradical activation is therefore not automatically equivalent to superior wastewater treatment.

4.4. Role of Co-PMS Complexes

An important development in the understanding of cobalt-mediated PMS activation is the recognition that Co-PMS complexes may participate directly in oxidation chemistry. Li et al. [9] provided evidence questioning whether freely diffusing radicals are necessarily the primary reactive species in all Co(II)/PMS systems and highlighted the role of a Co(II)-PMS complex. Mechanistic investigations of Co(II) complexes further demonstrated that PMS coordination and ligand environment can fundamentally affect reaction pathways [10]. The ligand or support environment surrounding cobalt can modify electron density, redox behavior, steric accessibility, and PMS-binding configuration. Coordination engineering may therefore change not only the rate of PMS activation but also the chemical nature of the dominant oxidizing pathway.

4.5. High-Valent Cobalt Species

High-valent cobalt species have been proposed as reactive intermediates in PMS activation. Such intermediates are mechanistically important because they could provide oxidation pathways distinct from conventional freely diffusing radicals. Their identification, however, remains challenging. Indirect probe reactions may not uniquely distinguish among Co(III), higher-valent cobalt species, radicals, and other reactive intermediates. Cao et al. [11] reinvestigated high-valent cobalt chemistry in the Co(II)/PMS system and emphasized the role of Co(III), demonstrating that some mechanistic assignments require reassessment. Accordingly, convincing identification of high-valent cobalt should ideally combine multiple independent approaches. Where direct identification remains incomplete, terminology such as “proposed

high-valent cobalt species" is scientifically preferable.

4.6. Coordination Environment as a Mechanistic Control Parameter

The local coordination environment provides the central connection between catalyst structure and PMS activation chemistry. A cobalt active site does not behave independently of its surroundings. The identity, number, geometry, and electronic properties of neighboring atoms influence cobalt electron density, orbital interactions, redox behavior, adsorption characteristics, and bond activation. Different coordination motifs, including Co-N, Co-O, and Co-Si environments, can therefore produce different interactions with PMS [8,15,16]. The structure-mechanism relationship can be expressed as: Coordination environment → cobalt electronic structure → PMS binding → electron transfer and peroxide activation → reactive intermediates → dominant oxidation pathway. Catalyst design should therefore move beyond simply maximizing cobalt loading toward controlling the chemical quality of cobalt active sites.

4.7. From Coordination Structure to Catalytic Selectivity

Coordination engineering can influence not only reaction rate but also catalytic selectivity. Changes in PMS adsorption geometry, electron-transfer direction, and intermediate stability can alter the balance among radical, nonradical, and surface-mediated pathways. This is particularly important for pharmaceutical contaminants because these compounds differ substantially in molecular structure, charge, electron density, functional groups, molecular size, and affinity for catalyst surfaces. Structure-activity relationships should consequently evolve toward structure-mechanism-selectivity relationships capable of explaining why particular coordination environments favor particular activation pathways and contaminant classes.

4.8. Standards of Mechanistic Evidence

One of the major challenges in Co/PMS research is distinguishing among simultaneously operating oxidation pathways. Radical-scavenging experiments are useful screening tools but should not be treated as definitive identification of reactive species. Stronger mechanistic assignments should combine complementary methods, including EPR, selective chemical probes, kinetic analysis, isotope-based experiments, electrochemical measurements, XAS-based characterization, in situ or operando spectroscopy, controlled catalyst comparisons, and computational calculations where appropriate. For heterogeneous systems, dissolved-cobalt measurements and solution-phase controls are also essential to distinguish genuine surface-mediated activation from homogeneous reactions caused by cobalt leaching. The strongest mechanistic argument therefore follows: Well-characterized active site + independently supported reactive pathway + consistent catalytic behavior.

V. Operational and Water-Matrix Factors Governing Co/PMS Performance

5.1. Effect of pH

Solution pH simultaneously affects cobalt speciation, PMS chemistry, catalyst surface charge, pharmaceutical ionization, and reactive-species behavior. In homogeneous systems, changes in pH can alter cobalt hydrolysis and precipitation. In heterogeneous systems, pH can modify catalyst-surface protonation and consequently affect adsorption of PMS and pharmaceutical molecules. A broad effective pH range is desirable from an engineering perspective because extensive pH adjustment increases chemical consumption and process complexity. Performance near the natural pH of wastewater may therefore be more practically significant than maximum reaction rates obtained under strongly optimized pH conditions.

5.2. Catalyst Loading

Increasing catalyst loading generally increases the number of accessible active sites and can accelerate PMS activation. The relationship, however, is not necessarily linear. Beyond an appropriate loading, additional catalyst may produce diminishing improvements when PMS or pollutant availability becomes limiting. Excess catalyst can also increase material consumption, separation requirements, and the total quantity of cobalt potentially available for leaching. The objective should therefore be efficient utilization of cobalt active sites rather than simply maximizing catalyst dosage.

5.3. PMS Dosage and Oxidant Utilization

Increasing PMS dosage can enhance pharmaceutical degradation when oxidant availability is limiting. Beyond an optimum, however, additional PMS may provide diminishing benefits, participate in nonproductive reactions, or remain as residual oxidant after treatment. Meaningful evaluation should therefore consider degradation and, where available, mineralization relative to PMS consumption. The focus should shift from maximum PMS dosage toward PMS-utilization efficiency.

5.4. Pollutant Identity and Concentration

Pharmaceuticals differ in molecular structure, charge, electron density, functional groups, hydrophobicity, molecular size, and affinity for catalyst surfaces. These differences affect their reactions with sulfate radicals, hydroxyl radicals, surface-bound oxidants, activated PMS complexes, and other reactive intermediates. High degradation efficiency toward a single model pharmaceutical therefore cannot establish broad-spectrum catalytic performance. Laboratory studies often employ concentrations substantially higher than those encountered in treated wastewater; such experiments remain valuable for mechanistic investigations but cannot independently establish performance at environmentally relevant trace concentrations.

5.5. Inorganic Ions

Real wastewater contains inorganic constituents such as chloride, bicarbonate, carbonate, phosphate, sulfate, and nitrate. These species may influence PMS chemistry through radical scavenging, secondary-reactive-species formation, surface interactions, competition for active sites, or modification of catalyst chemistry. The effect of an individual ion cannot be universally classified as inhibitory or beneficial because its influence depends on concentration, pH, catalyst structure, pollutant identity, and activation pathway.

5.6. Natural and Effluent Organic Matter

Natural organic matter and effluent organic matter can compete with pharmaceuticals for reactive species, consume oxidant, interact with PMS, and foul catalyst surfaces. Although some selective or surface-mediated pathways may be less susceptible to certain matrix components than highly reactive freely diffusing radicals, matrix resistance should be demonstrated experimentally rather than assumed from mechanistic classification.

5.7. Temperature, Reaction Time, and Variable Interactions

Increasing temperature can accelerate PMS activation, adsorption processes, and mass transfer but introduces additional energy requirements. Similarly, short reaction times can reduce reactor volume and improve treatment throughput, but rapid degradation may simply reflect excessive catalyst or oxidant input. Application-oriented optimization should therefore move beyond one-factor-at-a-time maximization toward multivariable and mechanistically informed evaluation.

5.8. Kinetic Interpretation

Pseudo-first-order kinetic models are frequently used to describe pharmaceutical degradation. Apparent rate constants can be useful for comparing conditions within the same experimental system. Direct comparison across different studies, however, requires caution because pollutant concentration, PMS dosage, catalyst loading, cobalt content, pH, temperature, reactor configuration, and water matrix can substantially affect reported kinetics. A larger apparent rate constant does not necessarily identify an intrinsically superior catalyst.

5.9. Integrated Treatment Performance

One of the most important limitations in the Co/PMS literature is the frequent use of parent-compound removal as the principal measure of catalytic success. A pharmaceutical can disappear rapidly while a substantial fraction of its organic carbon remains in solution as transformation products. High removal can also coexist with excessive PMS consumption or significant cobalt release. Parent-pharmaceutical disappearance ≠ complete mineralization ≠ demonstrated environmental safety. A stronger assessment integrates degradation, PMS utilization, mineralization, transformation products, cobalt leaching, catalyst stability, and post-treatment toxicity.

VI. Cobalt Catalyst Design and Active-Site Engineering

6.1. Design Requirements

A cobalt catalyst intended for wastewater treatment should ideally combine high activity, appropriate selectivity, efficient PMS utilization, structural stability, low cobalt leaching, recoverability, and reusability. These characteristics may involve trade-offs. Increasing cobalt accessibility may improve PMS activation while increasing dissolution risk; excessive stabilization may suppress cobalt leaching but reduce catalytic accessibility. Catalyst development should therefore be considered a multidimensional optimization problem rather than a search for maximum reaction rate.

6.2. Homogeneous versus Heterogeneous Cobalt Systems

Homogeneous Co(II) provides highly effective molecular-scale PMS activation [5,9,10]. However, dissolved cobalt remains in the treated aqueous phase unless an additional separation process is applied. Heterogeneous catalysts seek to retain cobalt within recoverable solids and potentially facilitate reuse. Nevertheless, a heterogeneous catalyst does not necessarily imply an exclusively

heterogeneous reaction mechanism or negligible cobalt leaching. Leached cobalt can contribute to PMS activation and confound both mechanistic interpretation and environmental assessment.

6.3. Role of the Catalyst Support

The support in a heterogeneous cobalt catalyst can perform functions extending beyond physical immobilization. It can influence cobalt dispersion, coordination geometry, electron density, surface area, porosity, PMS adsorption, contaminant adsorption, mass transfer, and metal-support electron transfer. In atomically dispersed catalysts, the support can effectively form part of the coordination environment of the cobalt center.

6.4. Engineering the Cobalt Coordination Environment

Coordination-environment engineering represents a transition from empirical catalyst formulation toward molecular-level active-site design. Rather than simply increasing cobalt loading, this strategy seeks to control the identity and arrangement of atoms directly interacting with cobalt. Co-N, Co-O, Co-Si, and other coordination environments can modify cobalt electron density, orbital interactions, redox behavior, and PMS-binding characteristics [8,15,16]. The central design question is which coordination environment provides the most favorable combination of PMS activation efficiency, pathway selectivity, cobalt stability, and wastewater-matrix tolerance.

6.5. Active-Site Density versus Active-Site Quality

Increasing the number of catalytic sites can increase overall reaction rate, but active-site density alone does not determine catalytic performance. Two cobalt centers with different coordination environments can exhibit substantially different electronic structures and PMS activation capabilities. Single-atom catalysts are particularly relevant because they can maximize metal utilization and allow comparatively precise manipulation of the local coordination environment. Their practical value, however, must still be judged against synthesis complexity, long-term stability, recovery, scalability, PMS demand, and real-wastewater performance.

6.6. Characterization of Cobalt Active Sites

A credible structure-mechanism relationship requires sufficiently rigorous characterization. XRD is valuable for identifying crystalline phases but provides limited information about highly dispersed or atomically isolated cobalt. SEM and TEM provide morphological and structural information, while BET analysis characterizes surface area and porosity. XPS can provide information about surface elemental composition and oxidation states but generally cannot independently establish a unique local coordination geometry or coordination number. When specific neighboring atoms, coordination numbers, or bond distances are central to the proposed mechanism, XANES and EXAFS can provide stronger evidence regarding the local cobalt environment.

6.7. Connecting Structure with Mechanism

Characterizing a cobalt coordination environment is not sufficient by itself. A meaningful structure-mechanism relationship requires evidence that modification of the identified coordination environment produces a corresponding and interpretable change in PMS interaction, reactive intermediates, kinetics, or product selectivity. Accordingly, the proposed structure-mechanism relationship should be evaluated through the following evidence-based sequence:

Verified active-site structure → demonstrated change in PMS interaction → independently supported activation mechanism → corresponding catalytic behavior

This sequence provides a more rigorous basis for establishing a causal structure-mechanism relationship and reduces the risk of attributing catalytic behavior to a proposed cobalt coordination environment solely on the basis of ex situ characterization or indirect mechanistic evidence.

6.8. Designing for Stability

Coordination engineering can potentially regulate both catalytic reactivity and cobalt retention. Strong metal-support or metal-ligand interactions may reduce dissolution, but excessive stabilization can compromise PMS interaction or necessary redox transitions. The desired condition is stable coordination without loss of catalytic accessibility.

VII. Environmental Stability, Cobalt Leaching, and Treatment Safety

7.1. Cobalt Leaching as a Critical Parameter

Cobalt leaching represents one of the principal limitations to practical Co/PMS application [12]. Exposure to PMS, changes in pH, repeated redox cycling, ligand interactions, and structural degradation can promote cobalt release from heterogeneous catalysts. Leaching is simultaneously an environmental and mechanistic issue. A process that removes a pharmaceutical while introducing substantial dissolved cobalt does not necessarily improve overall water quality. Furthermore, leached

cobalt may activate PMS homogeneously, causing apparent heterogeneous activity to be overestimated.

7.2. Controlling Cobalt Release

Approaches to suppress cobalt leaching include stronger metal-support interactions, structural confinement, encapsulation, surface modification, optimized catalyst composition, and coordination engineering [12]. The objective should not be maximum cobalt immobilization at any cost but an appropriate balance between catalytic accessibility and structural retention.

7.3. Reusability and Structural Retention

Repeated degradation cycles alone do not establish catalyst stability. Apparent activity can persist despite cobalt loss, surface reconstruction, oxidation-state changes, pore blockage, or contributions from dissolved cobalt. A rigorous reuse assessment should therefore consider activity retention, cobalt leaching, catalyst recovery, structural characterization before and after use, and preservation of the proposed coordination environment. Activity retention alone ≠ preservation of the active-site structure.

7.4. Parent-Compound Removal versus Mineralization

PMS oxidation can cause hydroxylation, dealkylation, deamination, ring opening, bond cleavage, and other structural transformations. The parent pharmaceutical may therefore disappear while a substantial fraction of its organic carbon remains in solution as transformation products. The treatment sequence should be distinguished as: Parent-compound removal → molecular transformation/degradation → mineralization. TOC or related measurements can provide information regarding the extent to which oxidation proceeds beyond initial molecular transformation.

7.5. Transformation Products

Identification of transformation products is important for understanding both chemical mechanisms and environmental consequences. High-resolution mass spectrometry can help identify intermediate products and support proposed degradation pathways. However, detection of an intermediate compatible with a proposed pathway does not necessarily establish the complete reaction sequence, particularly when multiple reactive species operate simultaneously.

7.6. Post-Treatment Toxicity

Transformation does not automatically eliminate biological activity. Oxidation products can have biological effects that differ from those of the parent compound. Where feasible, toxicity measurements should therefore complement chemical analysis. The preferred outcome is a simultaneous reduction in parent pharmaceutical, residual organic burden, and toxicity rather than merely a reduction in the analytical concentration of the original pharmaceutical.

7.7. Residual PMS and Final Water Quality

Residual oxidant is another important consideration. If PMS is supplied in substantial excess, unconsumed oxidant can remain after pharmaceutical degradation and may interfere with chemical or biological measurements. Relevant final-water endpoints include residual pharmaceuticals, transformation products, residual organic carbon, dissolved cobalt, residual PMS, and toxicity. Environmental safety should therefore be treated as an intrinsic component of catalytic performance.

VIII. Translation from Model Water to Real Wastewater

8.1. The Model-Water-to-Real-Wastewater Gap

Experiments in ultrapure or deionized water are valuable for isolating reaction mechanisms, but they represent only the first stage of technology evaluation. Real wastewater contains dissolved and particulate organic matter, inorganic ions, nutrients, multiple micropollutants, and other constituents that can consume PMS, quench reactive species, foul catalysts, alter adsorption, and modify cobalt speciation. High performance in ultrapure water does not necessarily predict high performance in real wastewater.

8.2. Matrix Competition and Oxidant Demand

In model solutions, the target pharmaceutical may represent one of the principal organic substrates available for oxidation. In real wastewater, pharmaceuticals coexist with a much larger background pool of organic and inorganic constituents. Reactive species generated through PMS activation can therefore be consumed by non-target substances, increasing oxidant demand and reducing the fraction of PMS effectively utilized for pharmaceutical degradation.

8.3. Environmentally Relevant Concentrations

Mechanistic experiments frequently employ pharmaceutical concentrations substantially higher than those encountered in treated wastewater because higher concentrations facilitate kinetic measurements and analytical detection. Such studies remain scientifically valuable but should not

automatically be extrapolated to trace-level treatment. Future application-oriented research should increasingly investigate lower concentrations and contaminant mixtures representative of actual wastewater.

8.4. Catalyst Separation and Recovery
Catalyst recovery is an essential engineering requirement for heterogeneous Co/PMS systems. A highly active catalyst that is difficult to separate from treated water can create operational challenges and potential release of cobalt-containing particles. Potential recovery approaches include filtration, membrane retention, magnetic separation where appropriate, sedimentation, and immobilized reactor configurations.

8.5. Long-Term Operation
Short reuse experiments in model solutions may underestimate deactivation under realistic conditions. Wastewater constituents can adsorb onto catalyst surfaces, occupy active sites, alter surface chemistry, or contribute to fouling. Application-oriented research should therefore progress toward longer-term repeated or continuous operation capable of detecting gradual cobalt loss, structural transformation, and catalyst fouling.

8.6. Coordination Engineering under Realistic Conditions
Studies evaluating coordination-controlled cobalt catalysts in real wastewater are particularly important because they connect molecular active-site design with practical treatment behavior [8,15]. If modification of the cobalt coordination environment improves performance not only in simplified solutions but also in complex wastewater, the evidence for engineering relevance becomes substantially stronger. Real-water validation should therefore become an integral stage of coordination-engineered catalyst development.

8.7. Application-Oriented Evaluation
A robust assessment of Co/PMS treatment in real wastewater should report, where applicable, wastewater characteristics; target pharmaceutical concentrations; catalyst dosage and cobalt input; PMS dosage and residual oxidant; reaction time or hydraulic residence time; parent-compound degradation; PMS consumption or utilization efficiency; mineralization or TOC reduction; transformation products; cobalt leaching; catalyst separation and recovery; catalyst reuse and structural stability; post-treatment toxicity; energy and chemical requirements; and preliminary economic considerations where sufficient data are available. Practical applicability can therefore be conceptualized as chemical efficiency + PMS efficiency + catalyst stability + effluent safety + catalyst recovery + reuse + operational feasibility.

IX. Comparative Analysis of Cobalt-Based PMS Activation Systems

The rapid development of cobalt-based PMS catalysts has produced systems ranging from homogeneous Co(II) to cobalt oxides, supported cobalt materials, heteroatom-coordinated sites, and atomically dispersed cobalt catalysts. Direct comparison is difficult because reported performance depends strongly on pollutant identity and concentration, catalyst dosage, PMS concentration, pH, reaction time, temperature, and water-matrix composition. Comparison based solely on pollutant-removal percentage can therefore be misleading. A more informative approach examines active-site structure, PMS activation mechanism, and environmental and practical performance.

Table 1. Comparative Features of Representative Cobalt-Based PMS Activation Systems

System	Cobalt Environment	Principal Mechanistic Significance	Treatment-Relevant Consideration	Reference
Homogeneous Co(II)/PMS	Dissolved Co(II)	Co-PMS complex and radical chemistry may coexist	Strong mechanistic benchmark; dissolved cobalt requires management after treatment	[9]
Co(II)-complex/PMS	Ligand-controlled cobalt environment	Ligand coordination can alter PMS interaction and reaction sequence	Highlights the importance of cobalt coordination in homogeneous systems	[10]
Co-Si coordinated catalyst/PMS	Co-Si local coordination	Coordination-induced nonradical PMS activation	Potential relevance of coordination control under real-wastewater conditions	[8]

Co single-atom catalyst on graphene-oxide-based support	Support-modulated isolated Co environment	Co/GO favored a proposed high-valent Co-oxo pathway; XAFS, in situ Raman, and DFT supported the coordination-reactivity relationship.	SMX mineralization reached 36.4% after 60 min; membrane configuration was also evaluated in real municipal wastewater.	[15]
Co single-atom catalyst on silica-based support	Support-modulated isolated Co environment	Co/SiO2 favored surface-bound radicals; the higher coordination number and stronger metal-support interaction were associated with the divergent pathway.	SMX mineralization reached 42.7% after 60 min; lower Co leaching and 7-day continuous real-wastewater membrane operation were reported.	[15]

Note: Direct numerical comparison among these systems should be made cautiously because pollutant concentration, catalyst loading, PMS dosage, pH, reaction time, reactor configuration, and water matrix differ among studies.

For the 2026 single-atom comparison, the reported mineralization values and continuous-operation observations are included because they were directly verified from the original study [15]; they should not be interpreted as universal benchmarks for other Co/PMS systems.

The comparative evidence supports several conclusions. First, catalytic performance cannot be explained solely by total cobalt content. The chemical state and local coordination environment of cobalt can strongly influence PMS interaction and activation [8,15,16].

Second, the classical radical-centered model is insufficient to describe the full range of cobalt-mediated PMS chemistry. Co-PMS complexes, surface-mediated processes, nonradical pathways, and cobalt species in different oxidation states must also be considered [6,9–11].

Third, coordination engineering provides a rational strategy for controlling catalytic behavior, but mechanistic novelty alone does not establish treatment superiority. A coordination-modified catalyst should ultimately demonstrate a measurable engineering advantage, such as improved PMS utilization, enhanced matrix tolerance, increased mineralization, reduced cobalt leaching, greater structural stability, or improved performance in real wastewater.

Finally, the most promising catalyst is not necessarily the material producing the fastest disappearance of a model pharmaceutical. The relevant comparison is: degradation efficiency + PMS utilization + mineralization + catalyst stability + low cobalt leaching + reusability + matrix tolerance + environmental safety.

X. Critical Synthesis: Major Gaps in Current Co/PMS Research

10.1. Structure-Mechanism Gap

A central limitation is the incomplete understanding of how specific cobalt coordination environments determine dominant PMS activation pathways. Many studies characterize catalyst composition, morphology, surface area, and oxidation state and subsequently propose mechanisms primarily from scavenger experiments or indirect reactive-species measurements. Such evidence does not necessarily establish a causal relationship between a particular cobalt coordination structure and a specific activation pathway. A stronger relationship requires: Local coordination environment → cobalt electronic structure → PMS binding configuration → electron transfer → reactive intermediate → dominant oxidation pathway. Recent coordination-controlled and single-atom studies provide important progress toward this goal [8,15,16]. Greater use of in situ and operando characterization is needed because the active structure under PMS exposure may differ from the as-synthesized material.

10.2. Mechanism-Performance Gap

Even when an activation mechanism is reasonably supported, its practical significance is not always demonstrated. The critical engineering question is whether the proposed mechanism

provides a measurable treatment advantage. A nonradical pathway may be mechanistically interesting but has limited practical value if it requires excessive PMS, produces poor mineralization, or suffers from cobalt instability. Conversely, a conventional radical pathway should not be considered inferior merely because it is less mechanistically novel if it provides efficient treatment at low chemical input with acceptable stability and safety.

10.3. Performance-Safety Gap

High pharmaceutical removal is frequently used as the principal measure of treatment success. However, parent-pharmaceutical disappearance ≠ complete mineralization ≠ demonstrated environmental safety. A catalyst can achieve near-complete parent-compound removal while leaving transformation products in solution, releasing cobalt, or leaving residual PMS. The relevant endpoint should therefore be final treated-water quality: parent removal → transformation products → mineralization → cobalt residuals → residual oxidant → toxicity.

10.4. Laboratory-Application Gap

Single-contaminant, ultrapure-water, optimized-pH batch experiments are appropriate for fundamental research but insufficient to establish practical applicability. Technology development should progress through: Mechanistic model system → complex synthetic matrix → real wastewater → repeated/continuous operation → process-level evaluation.

10.5. Catalyst-Stability Gap

Repeated-cycle degradation tests are frequently used to demonstrate reusability, but retained removal efficiency does not necessarily demonstrate preservation of the original active-site structure. This distinction is especially important for coordination-engineered catalysts. If the catalytic mechanism is attributed to a specific coordination environment, persistence of that environment during repeated PMS activation should be demonstrated where possible.

XI. Research Gaps and Future Directions

11.1. Greater Validation in Real Wastewater

Future studies should increase the use of real municipal, hospital, pharmaceutical, and other relevant wastewater matrices. Real-wastewater studies should report sufficient physicochemical characterization to allow matrix effects on PMS activation and catalyst stability to be interpreted.

11.2. More Rigorous Mechanistic Verification

Mechanistic assignments should increasingly rely on multiple complementary approaches rather than a single scavenger experiment. Greater integration of EPR, selective probes, isotope-based approaches, electrochemical analysis, advanced spectroscopy, kinetic modeling, and theoretical calculations would improve mechanistic confidence. Particular caution is required when assigning singlet oxygen, high-valent cobalt species, or surface-bound intermediates [9–11].

11.3. Rational Engineering of Cobalt Active Sites

Future catalyst development should shift further from empirical material screening toward rational control of the cobalt coordination environment. The key objective is to determine how coordination number, neighboring atoms, geometry, electronic structure, and support interactions influence PMS adsorption and activation [15,16].

11.4. Cobalt Leaching as a Design Constraint

Cobalt leaching should be considered during catalyst design rather than evaluated only after optimization. Future studies should investigate how coordination chemistry, support interactions, structural confinement, and surface modification can suppress cobalt dissolution without compromising PMS activation [12].

11.5. Pharmaceutical Mixtures

Most mechanistic studies employ a single target compound, whereas wastewater contains complex mixtures of pharmaceuticals and other micropollutants. Future research should examine competition for reactive species and catalyst surfaces and determine whether coordination-controlled activation can provide useful selectivity in multicomponent systems.

11.6. Transformation Products, Mineralization, and Toxicity

Greater attention should be given to the fate of organic carbon after disappearance of the parent pharmaceutical. Future studies should combine degradation measurements with TOC analysis, transformation-product identification, and appropriate toxicity assessment where feasible.

11.7. Locally Relevant Wastewater Conditions

Application-oriented research should consider the characteristics of the wastewater for which the technology is intended. For Kuwait and other water-stressed regions, this is particularly relevant. Evidence of pharmaceutical residues in Kuwaiti wastewater reinforces the need for locally relevant evaluation [4]. Assumptions regarding chloride concentration, ionic strength, organic matter, or other matrix characteristics should not replace direct measurement.

11.8. Economic and Sustainability Assessment

Catalyst performance should ultimately be considered together with material, chemical, and energy requirements. Relevant factors include catalyst-synthesis complexity, cobalt utilization, PMS consumption, catalyst lifetime, recovery requirements, energy demand, secondary-waste generation, and post-treatment requirements. The most promising catalyst is not necessarily the most structurally sophisticated material, but the system that provides the greatest overall treatment benefit with acceptable resource consumption, environmental impact, and operational complexity.

XII. Proposed Integrated Framework for Evaluating Co/PMS Catalysts

12.1. Level 1: Active-Site Structure

The first level establishes the structural identity of the cobalt active site. Relevant parameters include cobalt oxidation state, coordination number, neighboring atoms, bond distances, coordination geometry, dispersion, and support interactions. Key question: What is the chemically relevant cobalt site responsible for PMS activation?

12.2. Level 2: PMS Activation Mechanism

The second level determines how the characterized cobalt site interacts with PMS. Potential pathways include free-radical generation, surface-mediated activation, interfacial electron transfer, reactive Co-PMS complexes, proposed high-valent cobalt intermediates, and other nonradical processes [6,9–11]. Key question: How does the identified cobalt coordination environment alter PMS binding, electron transfer, and reactive-intermediate formation?

12.3. Level 3: Chemical Treatment Performance

Relevant indicators include degradation kinetics, parent-compound removal, PMS consumption, oxidant-utilization efficiency, selectivity, mineralization, and resistance to water-matrix interference. Key question: Does the identified activation pathway provide a measurable chemical-treatment advantage?

12.4. Level 4: Catalyst Stability

Relevant parameters include cobalt leaching, catalyst recovery, activity retention, structural changes after reaction, preservation of the coordination environment, and repeated-use performance. Key question: Does the cobalt active site remain chemically and structurally stable during PMS activation and reuse?

12.5. Level 5: Environmental Safety

Relevant endpoints include transformation products, mineralization, residual cobalt, residual PMS, and post-treatment toxicity. Parent-pharmaceutical disappearance ≠ complete mineralization ≠ demonstrated environmental safety. Key question: Does treatment reduce the overall chemical and biological burden of the water rather than merely remove the analytical signal of the parent pharmaceutical?

12.6. Level 6: Practical Applicability

Relevant factors include performance in actual wastewater, catalyst separation and recovery, repeated or continuous operation, chemical consumption, PMS efficiency, operating pH, reaction time, energy requirements, catalyst-synthesis complexity, scalability, and preliminary economic feasibility. Key question: Can molecular- and laboratory-scale advantages be retained under realistic treatment conditions?

12.7. Integration of the Six Levels

The complete framework is: Active-site structure → activation mechanism → chemical performance → catalyst stability → environmental safety → practical applicability. These levels should be treated as a causal sequence through which molecular-scale catalyst properties ultimately determine process-scale treatment performance. A weakness at any stage limits the strength of the overall conclusion.

XIII. Conclusions

Pharmaceutical residues remain an important wastewater-treatment challenge because conventional processes do not consistently eliminate all pharmaceutical micropollutants. PMS-based advanced oxidation offers a promising treatment strategy, and cobalt remains one of the most effective transition metals for PMS activation [1–5].

Nevertheless, contemporary evidence demonstrates that Co/PMS chemistry is substantially more complex than a universal Co(II)/Co(III) sulfate-radical cycle. Radical pathways remain important, yet Co-PMS complexes, surface-mediated species, electron-transfer processes, nonradical pathways, and cobalt species in different oxidation states can contribute under different chemical and structural conditions [6,9–11].

The coordination environment of cobalt therefore emerges as a central mechanistic control parameter. Neighboring atoms, coordination geometry, support interactions, and electronic structure can modify PMS adsorption, electron transfer, peroxide activation, and the resulting oxidation pathway [8,15,16]. The fundamental relationship can be summarized as: Cobalt coordination environment → electronic structure → PMS activation mechanism → catalytic behavior.

However, molecular-level control of PMS activation is not the final objective of water-treatment catalyst development. Its value depends

on whether it translates into improved PMS utilization, contaminant degradation, mineralization, matrix tolerance, cobalt retention, catalyst reuse, and final effluent quality.

High catalytic activity alone is insufficient to establish superior treatment performance. Likewise, disappearance of the parent pharmaceutical cannot by itself demonstrate complete mineralization or environmental safety.

Future catalyst development should therefore move from the narrow objective of maximizing pollutant-removal efficiency toward engineering the cobalt coordination environment to achieve simultaneous catalytic efficiency, selectivity, structural stability, environmental safety, and practical applicability.

Within this framework, the best Co/PMS catalyst is not necessarily the material producing the fastest disappearance of a pharmaceutical. Rather, it is the system providing the most favorable balance among degradation efficiency, PMS utilization, mineralization, low cobalt leaching, structural stability, reusability, matrix tolerance, treated-water safety, and operational feasibility. Future progress will therefore depend on connecting atomic-scale coordination chemistry with process-scale wastewater-treatment outcomes.

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