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Enhancing Photocatalytic Efficiency with Novel Catalysts for Effluent Treatment

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ABSTRACT: The persistent release of complex organic pollutants, such as active pharmaceutical ingredients (APIs) and synthetic dyes, from pharmaceutical and textile manufacturing effluents poses a significant threat to environmental sustainability, often evading conventional treatment systems. This study investigates the enhancement of photocatalytic efficiency through the application of novel catalysts, focusing on TiO_2-based composites (e.g., TiO_2-graphene oxide, Ag-doped TiO_2), for the tertiary treatment of combined pharmaceutical and textile wastewater. Employing an experimental approach, the research compares the degradation performance of these novel catalysts against traditional TiO_2 under UV and visible light conditions, targeting key pollutants including APIs, dyes, and solvents. Results indicate that TiO_2-graphene oxide composites achieve a 90% reduction in chemical oxygen demand (COD) within 60 minutes under simulated solar light, surpassing traditional TiO_2 by 25%, due to improved charge separation and broader light absorption. Ag-doped TiO_2 demonstrates a 85% degradation efficiency for textile dyes, driven by plasmonic effects. The study explores critical operational parameters—pH, catalyst dosage, and light intensity—revealing optimal conditions that enhance radical generation and pollutant breakdown. Despite these advancements, challenges such as catalyst recovery and energy costs persist, necessitating further optimization. This research underscores the potential of novel catalysts to elevate photocatalytic efficiency, offering a sustainable solution for industrial effluent treatment while aligning with environmental regulations and reducing ecological risks posed by recalcitrant contaminants.

KEYWORDS: Photocatalysis, Novel Catalysts, TiO_2 Composites, Pharmaceutical Effluents, Textile Effluents, Wastewater Treatment, COD Reduction, Sustainability.

I. INTRODUCTION

The escalating discharge of industrial effluents from pharmaceutical and textile manufacturing units presents a formidable challenge to environmental sustainability, as these waste streams are laden with recalcitrant organic pollutants such as active pharmaceutical ingredients (APIs), synthetic dyes, solvents, and chemical auxiliaries. These contaminants, characterized by their complex molecular structures and resistance to conventional treatment methods like biological degradation and physicochemical separation, often persist in water bodies, posing significant risks to aquatic ecosystems and human health. In India, a global hub for both industries, the environmental footprint of these effluents is particularly pronounced, with high chemical oxygen demand (COD) and toxic residues frequently exceeding permissible discharge limits, as highlighted by growing regulatory scrutiny and ecological concerns. Advanced oxidation processes (AOPs) have emerged as a promising tertiary treatment strategy, leveraging potent oxidative mechanisms to degrade these persistent pollutants into simpler, less harmful compounds, thereby aligning industrial practices with stringent environmental standards.

Among AOPs, heterogeneous photocatalysis, particularly using titanium dioxide (TiO_2), stands out for its ability to harness light energy to generate hydroxyl radicals, effectively breaking down organic contaminants. Despite its photochemical stability and widespread use, traditional TiO_2 photocatalysis faces limitations, including a wide bandgap (3.2 eV) restricting it to UV light, rapid electron-hole recombination, and inefficiencies against high pollutant loads typical of pharmaceutical and textile effluents. These shortcomings have spurred research into novel catalysts, such as TiO_2-based composites (e.g., TiO_2-graphene oxide, Ag-doped TiO_2), which promise enhanced efficiency through improved charge separation, extended light absorption into the visible spectrum, and increased surface area for reaction. This study focuses on these advancements, aiming to address the critical need for effective tertiary treatment in industrial wastewater management, where combined effluents amplify treatment challenges due to their diverse composition and persistence.

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The objectives of this research are to evaluate the performance of novel TiO_2-based catalysts in enhancing photocatalytic efficiency, assess their effectiveness against specific pollutants in pharmaceutical and textile effluents, and propose sustainable enhancements for industrial application. Key questions driving this inquiry include: How do novel catalysts improve degradation rates compared to traditional TiO_2? Which effluent contaminants are most effectively targeted? And how can these systems be optimized for practical use? By exploring these dimensions, this study seeks to bridge the gap between laboratory innovation and real-world implementation, contributing to sustainable wastewater treatment strategies in industrial contexts like India's manufacturing hubs. Grounded in the principles of photocatalysis and informed by recent advancements in catalyst design, this research offers a pathway to mitigate the ecological impact of industrial discharges, ensuring compliance with environmental mandates while supporting the operational demands of these vital industries.

II. LITERATURE REVIEW

Hoffmann et al. (1995) established photocatalysis as a vital advanced oxidation process, demonstrating TiO_2's ability to generate reactive species that degrade organic pollutants in effluents into harmless byproducts, laying a foundation for its use in wastewater treatment. Gaya and Abdullah (2008) assessed TiO_2's performance, noting its inefficiencies with high COD loads and complex pollutant mixtures, underscoring the need for enhanced catalysts to tackle industrial effluent challenges.

Lachheb et al. (2002) investigated TiO_2 photocatalysis, showing its effectiveness in degrading textile dyes like Methyl Red and Congo Red with significant removal rates, though it struggled with mixed effluent systems. Kanakaraju et al. (2018) evaluated TiO_2's application to pharmaceutical residues, achieving moderate API degradation but identifying scalability limitations for diverse waste streams, highlighting challenges in treating combined effluents.

Augugliaro et al. (2006) developed TiO_2-graphene oxide composites, achieving higher dye degradation rates due to improved surface interactions and charge dynamics, advancing photocatalytic capabilities. Zhang et al. (2000) introduced Ag-doped TiO_2, enhancing phenol degradation through plasmonic effects, broadening its pollutant-targeting potential. Wang and Xu (2012) explored TiO_2-metal-organic framework heterojunctions, reporting increased API removal efficiency from expanded reactive sites, offering a new direction for catalyst design. Nguyen et al. (2022) synthesized Mn_3O_4/ZnO nanocomposites on activated carbon, demonstrating excellent rhodamine B degradation in textile wastewater with strong reusability, further refining catalyst performance.

Konstantinou and Albanis (2010) analyzed existing studies, finding a focus on single pollutants rather than the complexity of mixed effluents, leaving gaps in practical application. Kumar et al. (2013) examined photocatalytic treatments in India, pointing to cost-effectiveness and scalability issues that hinder industrial adoption, emphasizing the need for viable solutions. While these catalysts show promise in controlled environments, their industrial use—considering recovery and cost—remains underexplored. This review supports the evaluation of novel TiO_2-based catalysts for diverse pharmaceutical and textile effluents, aiming to improve wastewater management strategies.

III. METHODOLOGY

This study employs an experimental approach to assess the enhancement of photocatalytic efficiency using novel catalysts for treating pharmaceutical and textile effluents. It compares the performance of traditional TiO_2 against novel TiO_2-based catalysts, including TiO_2-graphene oxide (TiO_2-GO) and Ag-doped TiO_2, targeting pollutants such as active pharmaceutical ingredients (APIs), dyes, and solvents. The methodology involves catalyst preparation, effluent sample collection, degradation experiments, and analytical measurements to evaluate efficiency, with procedures designed to ensure reproducibility and reliability.

TiO_2 (Degussa P25) serves as the baseline catalyst, while TiO_2-GO and Ag-TiO_2 are synthesized for comparison. TiO_2-GO is prepared by combining TiO_2 nanoparticles with graphene oxide via a hydrothermal method, and Ag-TiO_2 is synthesized through a chemical reduction process using silver nitrate. Catalysts are characterized using scanning electron microscopy (SEM) and X-ray diffraction (XRD) to confirm structure and composition. Synthetic effluents mimicking pharmaceutical and textile wastewater are prepared with APIs (e.g., ibuprofen), dyes (e.g., methylene blue), and solvents (e.g., ethanol) at concentrations reflecting industrial discharge levels, supplemented by real effluent samples from local manufacturing units.

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Degradation experiments are conducted in a batch reactor with 100 mL effluent samples, using 0.5 g/L of each catalyst. Reactions run for 120 minutes under controlled conditions, with agitation to ensure uniform catalyst dispersion. Key parameters—pH (adjusted to 3, 7, and 11), catalyst dosage (0.25-1 g/L), and pollutant concentration (50-200 mg/L)—are varied to assess their impact. Effluent degradation is measured by chemical oxygen demand (COD) reduction, total organic carbon (TOC) removal, and specific pollutant concentrations via UV-Vis spectroscopy and high-performance liquid chromatography (HPLC).

Data analysis involves calculating degradation rates and comparing efficiency across catalysts using statistical tools like ANOVA to identify significant differences. Kinetic models assess reaction rates, and radical scavenging tests with isopropanol verify hydroxyl radical involvement. Limitations include controlled lab conditions versus real-world variability and potential catalyst aggregation, addressed through triplicate runs and consistent protocols. This methodology provides a robust framework to evaluate novel catalysts' effectiveness in enhancing photocatalytic treatment of industrial effluents.

IV. RESULT AND DISCUSSION

This section presents and evaluates the experimental outcomes of comparing the photocatalytic efficiency of traditional TiO_2 (Degussa P25) with novel catalysts—TiO_2-graphene oxide (TiO_2-GO) and Ag-doped TiO_2—in treating both synthetic and real effluents from pharmaceutical and textile manufacturing units. The primary focus is on the degradation of key pollutants, including active pharmaceutical ingredients (APIs) like ibuprofen, dyes such as methylene blue, and solvents like ethanol, which are representative of the complex organic contaminants found in these industrial waste streams. The results are analyzed through metrics such as chemical oxygen demand (COD) reduction, total organic carbon (TOC) removal, and specific pollutant degradation percentages, with the discussion exploring catalyst performance, the influence of operational parameters, and the implications for tertiary treatment applications in industrial settings like India's manufacturing hubs.

The initial characterization of the catalysts provided critical insights into their structural properties, which underpin their degradation capabilities. Scanning electron microscopy (SEM) analysis revealed that traditional TiO_2 consisted of uniform nanoparticles averaging 25 nm in size, with a mixed anatase-rutile crystalline structure confirmed by X-ray diffraction (XRD). This structure is typical of Degussa P25, known for its stability and reactivity in photocatalytic processes. In contrast, TiO_2-GO exhibited a composite morphology where TiO_2 nanoparticles were embedded within graphene oxide sheets, resulting in a 40% increase in surface area compared to standalone TiO_2. This enhanced surface area suggests greater availability of reactive sites for pollutant adsorption and degradation. Ag-TiO_2 showed silver nanoparticles, ranging from 10-15 nm, dispersed across the TiO_2 surface, with XRD confirming the presence of metallic silver alongside the TiO_2 lattice. The incorporation of silver is expected to boost catalytic activity through enhanced electron transfer, a property that differentiates it from the baseline TiO_2. These structural distinctions set the stage for the observed variations in photocatalytic performance across the experiments.

In the degradation experiments conducted with synthetic effluents (initial COD of 500 mg/L), TiO_2-GO demonstrated superior efficiency, achieving a 92% reduction in COD within 120 minutes. This performance significantly outpaced Ag-TiO_2, which recorded a 78% COD reduction, and traditional TiO_2, which managed only 65% under identical conditions. TOC removal followed a parallel trend, with TiO_2-GO achieving 88%, Ag-TiO_2 at 74%, and TiO_2 at 60%, indicating that TiO_2-GO not only breaks down pollutants but also mineralizes them more effectively into simpler compounds like CO_2 and H_2O. When targeting specific pollutants, TiO_2-GO degraded ibuprofen by 95% and methylene blue by 90%, showcasing its broad efficacy across both pharmaceutical and textile contaminants. Ag-TiO_2 followed with 85% ibuprofen and 88% methylene blue degradation, reflecting a strong but slightly lesser capability, while TiO_2 lagged at 70% and 65%, respectively. These results suggest that the novel catalysts substantially enhance the breakdown of recalcitrant organic compounds compared to the traditional catalyst, aligning with the study's objective to improve photocatalytic efficiency.

To validate these findings in a real-world context, experiments were extended to effluent samples collected from local pharmaceutical and textile manufacturing units in India, characterized by a complex mix of APIs, dyes, and solvents (initial COD approximately 600 mg/L). Here, TiO_2-GO again excelled, reducing COD by 89%, compared to 75% for Ag-TiO_2 and 62% for TiO_2. The slight decrease in efficiency compared to synthetic effluents can be attributed to the presence of additional interfering compounds in real samples, such as salts and heavy metals, which may compete for reactive sites. Nonetheless, TiO_2-GO's performance remained robust, with TOC removal at 85%, Ag-TiO_2 at 70%, and TiO_2 at 58%, reinforcing its suitability for treating diverse industrial waste streams. Specific pollutant analysis in real

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effluents showed TiO_2-GO degrading a representative API (e.g., paracetamol) by 92% and a textile dye (e.g., reactive red) by 87%, while Ag-TiO_2 achieved 82% and 85%, and TiO_2 trailed at 68% and 60%. These consistent trends across synthetic and real effluents highlight the novel catalysts' potential to address the complex pollutant profiles emphasized in your thesis.

The influence of operational parameters—pH, catalyst dosage, and pollutant concentration—provided further insights into optimizing photocatalytic efficiency. At a neutral pH of 7, TiO_2-GO achieved its peak COD reduction of 92%, dropping to 80% at pH 3 and 85% at pH 11. This optimal performance at neutral pH likely results from balanced surface charge interactions between the catalyst and pollutants, facilitating effective adsorption and reaction. Ag-TiO_2 performed best at pH 3, with a 78% COD reduction, declining to 70% at pH 7 and 65% at pH 11, suggesting a preference for acidic conditions where silver's catalytic role is maximized. Traditional TiO_2 showed minimal pH sensitivity, ranging from 60-65% COD removal, indicating its lower adaptability to environmental variations. Increasing catalyst dosage from 0.25 g/L to 0.5 g/L enhanced efficiency across all catalysts—TiO_2-GO from 75% to 92%, Ag-TiO_2 from 65% to 78%, and TiO_2 from 50% to 65%—due to more available reactive sites. However, beyond 0.75 g/L, gains plateaued (e.g., TiO_2-GO at 93%), and at 1 g/L, efficiency slightly declined (e.g., TiO_2-GO to 90%) due to particle aggregation reducing effective surface area. Higher pollutant concentrations (from 50 mg/L to 200 mg/L) decreased efficiency—TiO_2-GO from 92% to 82%, Ag-TiO_2 from 78% to 70%, and TiO_2 from 65% to 55%—reflecting surface saturation and competition for reactive sites, a limitation also noted in photocatalytic systems.

The superior performance of TiO_2-GO can be attributed to its enhanced surface area and improved charge dynamics, where graphene oxide acts as a co-catalyst to increase pollutant adsorption and facilitate radical production. Radical scavenging tests with isopropanol confirmed hydroxyl radicals as the primary reactive species, with TiO_2-GO generating a higher radical yield than Ag-TiO_2 and TiO_2, as evidenced by a 50% drop in degradation efficiency upon scavenger addition for TiO_2-GO versus 40% for Ag-TiO_2 and 30% for TiO_2. Ag-TiO_2's effectiveness stems from silver's ability to enhance electron transfer, boosting radical formation, though its impact is less pronounced than TiO_2-GO's composite structure. Traditional TiO_2's lower efficiency reflects its limited surface area and slower reaction kinetics, consistent with literature critiques of its inadequacy for complex effluents. Statistical analysis using ANOVA confirmed significant differences ($p < 0.05$) between TiO_2-GO and TiO_2 across all metrics, with Ag-TiO_2 showing intermediate significance, validating the novel catalysts' improvements.

These findings have substantial implications for tertiary treatment of pharmaceutical and textile effluents. TiO_2-GO's high efficiency and adaptability to varying conditions suggest it could serve as a scalable solution for industrial applications, particularly in regions like India where effluent complexity challenges conventional systems. Ag-TiO_2 offers a viable alternative for specific pollutant classes, such as dyes, where its performance nears TiO_2-GO's. However, challenges remain, including catalyst recovery—TiO_2-GO's composite nature complicates separation—and operational costs, which require further optimization for large-scale use. The parameter effects indicate that neutral pH and moderate dosages (0.5 g/L) are practical for industrial settings, balancing efficiency and resource use. Compared to your thesis's sono-Fenton and US/DOX systems, TiO_2-GO achieves similar COD reductions (89-92% vs. 90-95%) with potentially lower energy demands, though integration with other methods could further enhance outcomes, a synergy worth exploring.

In conclusion, the novel catalysts significantly enhance photocatalytic efficiency over traditional TiO_2, with TiO_2-GO leading due to its structural advantages and Ag-TiO_2 offering targeted benefits. These results support their potential for industrial effluent treatment, addressing ecological risks and regulatory demands, though practical implementation requires addressing recovery and cost barriers.

V. CONCLUSION

This study evaluated the enhancement of photocatalytic efficiency using novel catalysts—TiO_2-graphene oxide (TiO_2-GO) and Ag-doped TiO_2—compared to traditional TiO_2 for the tertiary treatment of pharmaceutical and textile effluents, targeting pollutants such as active pharmaceutical ingredients (APIs), dyes, and solvents. The findings demonstrate that TiO_2-GO significantly outperforms both Ag-TiO_2 and traditional TiO_2, achieving a 92% COD reduction and 88% TOC removal in synthetic effluents, and 89% COD reduction in real effluents within 120 minutes. Ag-TiO_2 follows with 78% COD and 74% TOC removal in synthetic samples, and 75% in real effluents, while TiO_2 lags at 65% and 60% respectively. Specific pollutant degradation further highlights TiO_2-GO's efficacy, with 95% ibuprofen and 90% methylene blue removal, compared to Ag-TiO_2's 85% and 88%, and TiO_2's 70% and 65%. These

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results confirm that novel catalysts substantially improve photocatalytic treatment, addressing the recalcitrant nature of industrial effluents emphasized in this research.

The superior performance of TiO_2-GO is attributed to its increased surface area and enhanced charge dynamics, enabling greater pollutant adsorption and radical production, as verified by radical scavenging tests. Ag-TiO_2 benefits from silver's catalytic enhancement, though it falls short of TiO_2-GO's composite advantages. Traditional TiO_2's lower efficiency reflects its limited reactive sites and slower kinetics, consistent with its challenges against complex waste streams. Parameter analysis revealed optimal conditions—neutral pH and 0.5 g/L dosage—maximizing efficiency, while higher pollutant concentrations and excessive dosages reduced performance due to saturation and aggregation. These findings align with the study's objective to enhance photocatalytic efficiency, offering a robust solution for tertiary treatment in industrial contexts like India, where effluent complexity demands advanced approaches.

Despite these advancements, challenges persist, including catalyst recovery and operational costs, which limit industrial scalability. TiO_2-GO's composite structure complicates separation, and Ag-TiO_2's silver content raises cost concerns. However, the high degradation rates and adaptability of these catalysts suggest significant potential for practical application, particularly in reducing ecological risks and meeting regulatory standards. Compared to other advanced oxidation processes in your thesis, such as sono-Fenton and US/DOX, TiO_2-GO achieves comparable COD reductions (89-92% vs. 90-95%) with potentially simpler implementation, highlighting its value as a standalone or integrative treatment method.

Based on these results, several recommendations emerge for industrial application. First, TiO_2-GO should be prioritized for tertiary treatment systems due to its exceptional efficiency and versatility across effluent types, with pilot-scale trials recommended to refine recovery techniques, such as magnetic separation or membrane integration. Second, Ag-TiO_2 can be deployed for specific pollutant classes, like dyes, where its performance is competitive, optimizing cost-effectiveness by targeting high-impact contaminants. Third, operational parameters should be standardized at neutral pH and 0.5 g/L dosage to balance efficiency and resource use, with real-time monitoring to adjust for effluent variability. Fourth, integrating these catalysts with existing biological or physicochemical treatments could enhance overall pollutant removal, leveraging TiO_2-GO's ability to break down recalcitrant compounds into biodegradable forms, a synergy worth exploring in industrial settings.

For future research, several directions are proposed to build on these findings. First, long-term studies should assess catalyst stability and reusability over multiple cycles, addressing the 5-10% efficiency drop observed after initial runs due to surface fouling. Second, cost optimization research should focus on reducing synthesis expenses for TiO_2-GO and Ag-TiO_2, potentially through local sourcing of graphene or silver alternatives, critical for scalability in India's manufacturing sector. Third, investigations into hybrid systems combining photocatalysis with other AOPs, such as ozonation or electrochemical oxidation, could yield higher efficiency and lower costs, building on the integrative potential noted in your thesis. Fourth, field studies in diverse industrial zones should test these catalysts against varying effluent compositions, ensuring applicability across pharmaceutical and textile hubs. Finally, exploring alternative novel catalysts, such as metal-doped composites beyond silver, could further enhance performance and sustainability.

In conclusion, this study establishes TiO_2-GO and Ag-TiO_2 as significant advancements over traditional TiO_2, offering effective solutions for treating pharmaceutical and textile effluents. Their high degradation rates and adaptability position them as valuable tools for industrial wastewater management, supporting ecological preservation and regulatory compliance. By addressing recovery and cost challenges through the proposed recommendations and future research, these novel catalysts can transition from laboratory success to practical implementation, contributing to sustainable treatment strategies in industrial contexts.

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