



The effect of microemulsion method on the characteristics of $\text{Fe}_3\text{O}_4/\text{TiO}_2$ material and its relationship with performance in methylene blue degradation: A Systematic Review

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Abstract

TiO_2 -based photocatalysis offers a potential solution, but it has limitations, including high electron-hole recombination and difficulty in separating the catalyst after the reaction. The development of $\text{Fe}_3\text{O}_4/\text{TiO}_2$ nanocomposites via the microemulsion method was conducted to enhance performance while facilitating catalyst separation. This study aims to examine the influence of the microemulsion method on material characteristics, characterization results, and their relationship to methylene blue degradation performance. The study was conducted using a systematic review with a PRISMA-compliant approach on literature from Scopus over the past 10 years. The selection process yielded five relevant articles. The microemulsion method was proven capable of producing nanoparticles (8–50 nm) with a homogeneous distribution and a mesoporous structure with a high surface area. XRD and SEM characterization results showed a dominance of the anatase phase or a mixture of anatase-rutile, which plays a role in enhancing charge separation. Photocatalytic performance showed a methylene blue degradation efficiency of up to approximately 90% within 60 minutes under sunlight. The improved performance was influenced by small particle size, high surface area, pore structure, and crystal phase composition. The addition of Fe_3O_4 has the potential to enhance electron transfer, suppress recombination, and facilitate catalyst separation through its magnetic properties. The microemulsion method plays a direct role in controlling the structure and characteristics of the material, which leads to improved photocatalytic performance of $\text{Fe}_3\text{O}_4/\text{TiO}_2$ in the degradation of methylene blue.

INTRODUCTION

Water pollution caused by synthetic dye waste is an increasingly complex and urgent environmental issue that requires attention [1]. The rapid growth of the textile, printing, paper, and pharmaceutical industries has increased the volume of liquid waste containing hazardous organic compounds, particularly synthetic dyes that are stable and difficult to break down naturally [2]. One of the most common dyes found in industrial wastewater is methylene blue, which belongs to the cationic dye group [3]. This compound has a complex aromatic structure that makes it resistant to biodegradation and has the potential to cause negative impacts on aquatic ecosystems and human health, such as skin irritation, respiratory problems, and toxic effects on aquatic organisms [4].

Various methods have been developed to address dye contamination in water, such as adsorption, coagulation-flocculation, chemical oxidation, and membrane filtration. Although these methods are quite effective in reducing dye concentrations, most merely transfer pollutants from one phase to another without actually breaking down their chemical structure. Therefore, it is crucial to develop technologies capable of completely degrading organic compounds [5]. One promising approach is advanced oxidation processes (AOPs), particularly photocatalysis, which utilizes hydroxyl radicals (OH) as a strong oxidizing agent to break down complex organic compounds into simpler, non-hazardous products [6]. Titanium dioxide (TiO₂) is the most extensively studied photocatalytic material for dye degradation applications due to its high chemical stability, non-toxicity, relatively low cost, and good photocatalytic activity under ultraviolet irradiation [7]. However, the use of TiO₂ as a single photocatalyst still faces several limitations, including a high rate of electron-hole pair recombination, limited active surface area, and difficulties in separating and recovering the catalyst from the solution after the process is complete [8]. These limitations have driven the development of composite materials capable of enhancing photocatalytic efficiency while simplifying the separation process [9].

One widely developed strategy involves combining TiO₂ with magnetic materials such as Fe₃O₄ (magnetite) [10]. Fe₃O₄ possesses strong magnetic properties, enabling easy separation of the catalyst using an external magnetic field [11]. Additionally, the presence of Fe₃O₄ in the composite can help enhance charge transfer and reduce the electron-hole recombination rate, thereby potentially increasing photocatalytic activity [12]. Thus, the Fe₃O₄/TiO₂ composite is a highly promising candidate material for dye degradation applications, including methylene blue [13]. The performance of Fe₃O₄/TiO₂ composites is significantly influenced by their physical and chemical characteristics, such as particle size, morphology, specific surface area, crystal structure, and the distribution and interactions between the Fe₃O₄ and TiO₂ phases [14]. These characteristics are fundamentally determined by the synthesis method employed. Therefore, selecting the appropriate synthesis method is a key factor in producing materials with optimal performance [15]. Various synthesis methods have been reported, such as sol-gel, coprecipitation, hydrothermal, and microemulsion, each with its own advantages and limitations [16].

The microemulsion method is an intriguing approach in the synthesis of nanocomposites due to its ability to control particle size and distribution with greater precision [16]. A microemulsion is a thermodynamically stable isotropic dispersion system consisting of an oil phase, an aqueous phase, a surfactant, and often a co-surfactant. In this system, nano-sized droplets form, functioning as “nanoreactors” where chemical reactions take place [17]. The extremely small and uniform droplet size allows for the formation of particles with controlled size, narrow distribution, and more homogeneous morphology compared to conventional methods [18]. The advantages of the microemulsion method lie not only in particle size control but also in its ability to facilitate better interaction between the Fe₃O₄ and TiO₂ phases. This is crucial because the synergy between these two components significantly determines charge transfer efficiency and photocatalytic activity [19]. Furthermore, parameters within the microemulsion system such as surfactant type, water-to-oil ratio, precursor concentration, and reaction conditions can be modified to produce materials with desired characteristics [20]. However, the complexity of the microemulsion system also presents a unique challenge, as even minor changes in synthesis parameters can result in significant differences in the properties of the resulting material [21].

Several studies have reported the successful synthesis of Fe₃O₄/TiO₂ composites using the microemulsion method under various synthesis conditions [22]. The results obtained indicate that this method is capable of producing nanoparticles with uniform morphology and high photocatalytic activity in the degradation of methylene blue [23]. However, there are significant variations in the reported characterization results and photocatalytic performance, attributed to differences in synthesis parameters and experimental conditions [24]. This indicates that the

relationship between the microemulsion method, material characteristics, and photocatalytic performance has not yet been fully and comprehensively understood [25].

Based on this, a systematic review is needed to integrate existing research findings to identify patterns, trends, and key factors influencing the success of synthesis and material performance. A systematic review approach is highly relevant in this context, as it enables a structured and comprehensive analysis of the available literature. Using this approach, an evaluation can be conducted of the influence of microemulsion methods on the characteristics of Fe₃O₄/TiO₂ materials, the characterization results obtained (such as XRD and SEM), and their relationship to performance in the degradation of methylene blue [26], [27]. Accordingly, the research question in this study is to determine how the microemulsion method affects the characteristics and characterization results of Fe₃O₄/TiO₂ materials, as well as their relationship to performance in the degradation of methylene blue. This research question emphasizes the importance of understanding the interrelationship between the synthesis process, material structure, and application performance, thereby providing a strong scientific foundation for the development of more effective photocatalytic materials.

Thus, the objective of this article is to conduct a systematic review of the synthesis of Fe₃O₄/TiO₂ composites using the microemulsion method, focusing on identifying synthesis parameters that influence material characteristics, analyzing characterization results reported in various studies, and evaluating the relationship between material characteristics and photocatalytic performance in the degradation of methylene blue.

METHOD

Research Design

This study employed a systematic literature review using a qualitative descriptive approach in accordance with the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) guidelines. The literature search was conducted using the Scopus database as the primary source and Google Scholar, with the assistance of the Publish or Perish tool, as a supplementary source. The keywords used were tailored to the research topics, namely the microemulsion method, the synthesis of Fe₃O₄ and TiO₂, and the degradation of methylene blue. The literature selection process was conducted in stages, including identification, screening, and eligibility assessment. In the initial stage, articles were filtered based on publication type, excluding review articles and book chapters, and limited to the past 10 years. Subsequently, the articles that passed were selected based on inclusion and exclusion criteria, including the availability of full-text access, topic relevance to the research, and relevance to the synthesis methods and material applications under study. Eligibility was assessed by reviewing the titles and abstracts. Articles that met the criteria were then analyzed using content analysis, focusing on microemulsion synthesis methods, the characteristics of Fe₃O₄ and TiO₂ materials, and their performance in the degradation of methylene blue. The data obtained were then presented descriptively in narrative form and tables to facilitate interpretation and the drawing of conclusions.

Data Sources and Literature Search Strategy

The data sources for this study consisted of scientific articles obtained from the Scopus database as the primary source, as well as Google Scholar via the Publish or Perish tool as a supplementary source. The literature search was conducted using keywords relevant to the research topic. The literature search strategy follows the PRISMA flowchart, which includes the identification and selection stages, as shown in Figure 1. The retrieved articles were then filtered based on publication type, excluding review articles and book chapters, and limited to publications from the past 10 years. Subsequently, only articles available in open access and fully downloadable were selected. The next stage involved selection based on topic relevance through a review of titles and abstracts. At this stage, inclusion and exclusion criteria were applied to eliminate irrelevant articles, resulting in a set of articles suitable for further analysis.

Article retrieval was conducted through several selection stages. Complete data, along with the keywords used in the search process, are presented in Table 1.

Table 1. Sources, Boolean Search, Include and Exclude

| Database | Bolean search |
|----------------|--|
| Google Scholar | : ("micro emulsion") AND ("TiO ₂ " AND " Fe ₃ O ₄ ") AND ("Methylene Blue" OR "Textile") AND ("Nanocomposite")) AND ("methylene blue") AND (microemulsion OR "micro-emulsion" |
| Scopus | : Fe ₃ O ₄ " OR magnetite OR " TiO ₂ " OR "titanium dioxide") AND ("methylene blue") AND (microemulsion OR "micro-emulsion" |

Inclusion and Exclusion Criteria

The inclusion criteria in this study comprised articles that specifically discussed the use of microemulsion methods in the synthesis of Fe₃O₄ and TiO₂, as well as studies evaluating the application of these materials in the degradation of Methylene Blue dye. Only publications released within the last ten years were considered to ensure the relevance and novelty of the collected data. In addition, articles had to match the research scope based on the results of the Boolean search strategy. Meanwhile, the exclusion criteria included articles that were not available through open-access sources, studies that were not relevant to the research theme based on title and/or abstract screening, review papers, and book chapters.

Study Selection Procedure

The literature selection process was conducted systematically in accordance with the PRISMA 2020 flowchart, which comprises four main stages, identification, screening, eligibility assessment, and final inclusion.

1. Identification: All articles obtained from the Google Scholar and Scopus databases were collected and documented as initial data.
2. Screening: Initial selection was based on titles and abstracts. Articles not relevant to the research focus, namely, microemulsion methods, the synthesis of Fe₃O₄ and TiO₂, and their application in the degradation of methylene blue dye, were eliminated at this stage.
3. Eligibility: Articles that pass the screening stage are then thoroughly analyzed through a full-text review. The evaluation is conducted by considering all inclusion and exclusion criteria, including the type of publication and the relevance of the content to the research topic. Articles that do not meet the requirements will be excluded from the selection process.
4. Inclusion: Articles that meet all criteria are then designated as the final sample. These articles serve as the basis for analysis, particularly to assess the performance of the material in the methylene blue degradation process.

This series of steps aims to ensure that the literature used is of sufficient quality and highly relevant to the research topic.

Data Extraction

Data extraction was performed on articles that met the inclusion criteria and passed all stages of the PRISMA selection process. This process aimed to systematically identify and collect key information from each article. The extracted data included the authors' names and year of publication, the synthesis methods used (specifically the microemulsion method), the types of materials synthesized (Fe₃O₄ and/or TiO₂), synthesis conditions, and the material characterization performed. Additionally, data regarding the application of the materials in the degradation of methylene blue dye were extracted, such as testing conditions (light source, reaction time, and initial concentration), as well as the resulting degradation efficiency. Additional information, such as the advantages of the method, factors affecting performance, and the main research results, was also recorded to support the analysis. All the data obtained was then organized into summary tables to facilitate the process of comparing articles and drawing conclusions.

Data Synthesis and Analysis

Data synthesis was conducted by combining and comparing findings from selected articles to obtain an overview of the use of the microemulsion method in the synthesis of Fe₃O₄ and TiO₂ and its application in the degradation of methylene blue. The synthesis process focused on the similarities and differences in synthesis methods, reaction conditions, and the characteristics of the resulting materials. Additionally, the effectiveness of the materials in the degradation process was compared based on parameters such as reaction time, light source, and degradation efficiency. Data analysis was conducted using qualitative descriptive methods with content analysis techniques. Each article was analyzed to identify the relationship between microemulsion synthesis methods and the properties and performance of the materials in methylene blue degradation. The analysis results were then interpreted to determine the factors influencing degradation effectiveness, such as particle size, surface area, and material interactions within the composite system. Thus, this analysis provides insight into the advantages of the microemulsion method in enhancing material performance as a dye degradation agent.

RESULT AND DISCUSSION

Article Selection Using the PRISMA Flowchart

Articles were selected following the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) flowchart, as shown in Figure 1.

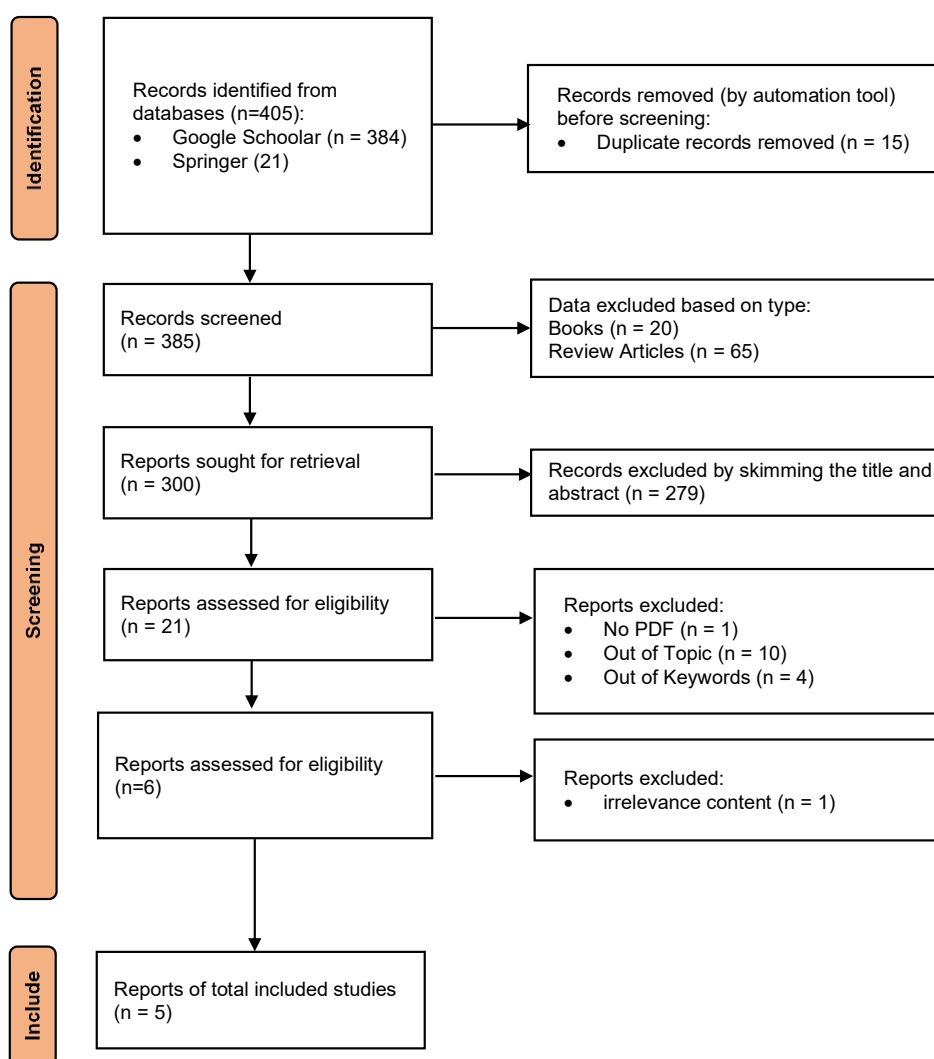


Figure 1. PRISMA Results

Based on this workflow, the identification phase using the Publish or Perish tool with Google Scholar as the source yielded 384 articles, while a search in the Scopus database yielded 21 articles. The primary source used in this study was Scopus. From the 21 articles obtained, a selection process was conducted by excluding review articles and book chapters, and limiting the publication timeframe to the last 10 years. The selection results showed that only 6 articles were available in open access and could be downloaded. Subsequently, these six articles were further screened based on their relevance to the research theme, namely microemulsion methods, the synthesis of Fe₃O₄ and TiO₂, and their application in the degradation of methylene blue dye. The selection process involved reviewing titles and abstracts to eliminate irrelevant articles. Some inclusion and exclusion criteria were also applied at this stage. Based on this entire process, five articles meeting the criteria were identified and subsequently used for further analysis.

Literature Review Article Profile

Table 2 shows that TiO₂-based research is advancing in two main directions, energy applications and photocatalysis, using a variety of synthesis approaches. A study by Zhang et al. [28] in China highlights the use of the microemulsion electrospinning method to produce hierarchically porous TiO₂ nanofiber structures intended for use as lithium-ion battery anodes; this study also has the highest number of citations (38), indicating its significant influence in the field. Meanwhile, more recent research from Italy, such as that by Marconi et al. and Marconi & Tortora, focuses on microemulsion-based synthesis using templating and water-in-oil (W/O) approaches, which tend to be more environmentally friendly (green approaches) to enhance photocatalytic activity, although their citation counts remain low (6 and 2) due to their relative novelty [29], [30]. On the other hand, research by Honarmand et al. [31] from Iran emphasizes the modification of TiO₂ through zirconium (Zr) doping using the microemulsion method to enhance photocatalytic activity in the degradation of methylene blue, with a moderate number of citations (23) indicating fairly strong relevance. A similar approach is also seen in the study by Rosa et al. from Italy, which uses iron (Fe) doping via the solid-state method for the purpose of dye degradation, with 14 citations [32].

Table 2. Literature Review Article Profile

| Ref. | Country | Journal Title | Publisher | Keywords | Cit. |
|------|---------|--|--------------------|--|------|
| [28] | China | BEILSTEIN Journal of Nanotechnology | Beilstein-Institut | anode; hierarchically porous TiO ₂ nanofibers; lithium-ion batteries; microemulsion electrospinning (ME-ES); multichannel | 38 |
| [29] | Italia | Acta IMEKO | IMEKO | Titanium dioxide; microemulsion; templating synthesis; photocatalysis | 6 |
| [31] | Iran | SN Applied Sciences | Springer Nature | TiO ₂ ; Zr-doped TiO ₂ ; Surfactants; Microemulsion method; Photocatalytic activity; Methylene blue | 23 |
| [32] | Italia | Journal of Materials Research | Springer Nature | Photocatalysis, Visible light, Nanoparticles, Methylene blue, Doping | 14 |
| [30] | Italia | 2023 IMEKO TC-4 International Conference | IMEKO | Titania nanoparticles; W/O microemulsion; green approach; photoactivity | 2 |

Based on Table 2, the keywords that emerge indicate a focus on improving material performance through structural engineering (porosity, morphology) and composition (metal doping), as well as their application in waste treatment and energy. The distribution of publications is dominated by Italy and supported by major publishers such as Springer Nature and scientific organizations such as IMEKO, indicating a concentration of research at specific institutions. Variations in citation counts also indicate that older research has a more established impact, while recent research is still in the stages of development and scientific dissemination.

Parameters Analyzed in Microemulsions

Table 3 shows that the microemulsion methods used vary, particularly between water-in-oil (W/O) systems and general microemulsions, with one specific approach being microemulsion electrospinning (ME-ES). The phase ratio (Rv/Rw) is not always reported consistently; however, in some studies, such as those by Zhang et al. and Marconi et al. [28], [29], the ratio is strictly controlled because it affects droplet stability and the resulting particle size. The types of surfactants used include cationic (CTAB, CPB), anionic (AOT), and nonionic (TX-100), and there are even combinations of several surfactants in a single system. This indicates that surfactant selection is tailored to the stability requirements of the microemulsion and its interaction with the precursor.

Table 3. Summary of Microemulsion Parameters

| Ref | R _v /R _w | Type | Surfactant | Co-surfact. | Oil Phase | Visco. |
|------|---|--|---|-------------------|---|-------------------------|
| [28] | Microemulsion electrospinning (ME-ES) | TBT/paraffin oil 2,25; 1.9; 1.55 (w/w) | CTAB (cetyltrimethylammonium bromide) | 0.3 g | Butoxy group in TBT (as an additional surfactant) | Paraffin oil |
| [29] | Water-in-oil (W/O) | Rasio volume 1:1 | CPB (cetylpyridinium bromide) | 1,0 g | 1-pentanol (1,5 mL) | Cyclohexane (30,0 mL) |
| [31] | Microemulsion method | NR | AOT (anionik), TBAB (kationik), TX-100 (nonionik) | 0.15 M and 0,08 M | Butanol | Ethanol (fase kontinyu) |
| [32] | Microemulsion method (sTiO ₂) | Ethanol (20 mL) : Air (40 mL) | NR | NR | NR | Ethanol |
| [30] | Water-in-oil (W/O) | Rasio volume 1:1 | CPB (cetylpyridinium bromide) | 1.0 g | 1-pentanol (1,5 mL) | Cyclohexane (30,0 mL) |

Note: NR is Not Reported

Surfactant concentrations also vary (from grams to molar), indicating different approaches to controlling micelle size and particle distribution. The use of co-surfactants such as butanol and 1-pentanol plays a role in reducing interfacial tension and increasing the flexibility of the surfactant layer, thereby aiding in the formation of stable microemulsions. The oil phase typically consists of nonpolar solvents such as paraffin oil and cyclohexane, although some studies use ethanol as the continuous phase, indicating a shift toward semi-polar systems. Emulsification conditions are characterized by prolonged stirring (4–24 hours) and, in some cases, followed by high-temperature reflux (70–80 °C).

Material Characteristics

The material characteristics of TiO₂ are shown in Table 4. Table 4 shows that the characteristics of TiO₂ materials are significantly influenced by the synthesis method, thermal conditions, and compositional modifications. A study by Zhang et al. produced hierarchically porous TiO₂ nanofibers via microemulsion electrospinning with relatively small particle sizes (17.8–20.1 nm) and a uniform meso-macro pore distribution, as well as a stable tetragonal anatase phase upon calcination at 500°C [28]. A microemulsion-based approach was also used by Marconi et al. and Marconi & Tortora [29], [30], but resulted in larger aggregates (~900 nm) with smaller crystallite sizes (8–50 nm) and high surface areas (250–256 m²/g), and showed the formation of a mixed anatase-rutile phase (both of which possess tetragonal crystal structures) due to variations in calcination temperature (450–600 °C). Modification via doping has a significant effect on particle size, structure, and porosity properties. Research by Honarmand et al. showed that Zr doping aided by surfactants in a microemulsion system can produce

spherical particles of 40–50 nm in size with a more homogeneous distribution and prevent agglomeration [31], while maintaining the dominance of the anatase phase with a tetragonal crystal structure. Meanwhile, the study by Rosa et al. showed that Fe doping via the solid-state method can reduce particle size (from 62.76 to 39.88 nm) and increase surface area and mesopores [32], while simultaneously inducing a phase transformation into an anatase–rutile mixture with tetragonal crystal structures.

Table 4. Material characteristics of TiO₂

| Ref. | Country | Composite material | Synthetic method | Calcination temp. | Drying Cond. | Particle size | Porosity | Phase & Structure |
|------|---------|---|---|-------------------|---|--|---|---|
| [28] | China | Hierarchically porous TiO ₂ nanofibers | Microemulsion electros pinning | 500 °C | 60 °C 12 hours in a vacuum oven | 17,8-20,1 nm | Pores 17.8–20.1 nm (meso-macropores) | Anatase; Tetragonal |
| [29] | Italia | TiO ₂ nanoparticles | Microemulsion (W/O 1:1) and one-pot | 450 & 600 °C | NR | Aggregates ~900 nm; grains 30-50 nm | ~250 m ² /g; mesopores | Anatase + Rutile (mixture); Tetragonal |
| [31] | Iran | Zr-doped TiO ₂ | Microemulsion | 500 °C | 70 °C for 12 hours in an oven to remove the solvent | 40-50 nm (spherical) | Surfactant micelles prevent agglomeration; uniform distribution | Anatase (surfactant); Tetragonal (Zr-doped) |
| [32] | Italia | Fe-doped TiO ₂ | Solid-state (using microemulsion-derived titania) | 600 °C | 60 °C overnight in an oven | 62.76 to 39.88 nm (0.5% Fe) | Mesopores; surface area increases with the addition of Fe | Anatase + Rutile (Fe-induced); Tetragonal |
| [30] | Italia | Titania (TiO ₂) nanoparticles | Microemulsion (W/O 1:1) | 450 & 600 °C | NR | Aggregates ~900 nm; crystallites 8-10 nm | Mesopores; surface area up to 256 m ² /g | Anatase + Rutile (Fe-induced); Tetragonal |

Application to Methylene Blue

The characteristics of TiO₂ in degrading methylene blue are shown in Table 5. Table 5 shows that the degradation performance of methylene blue is significantly influenced by the type of material, operating conditions, and the light source used. In this study, degradation efficiency refers to the percentage reduction of methylene blue concentration relative to its initial concentration, indicating the effectiveness of the photocatalytic process. The comparison of degradation time and efficiency shows that higher efficiency can be achieved in shorter time when the photocatalyst system is more effective, while longer reaction time does not necessarily result in significantly higher efficiency. The TiO₂-based microemulsion materials from Marconi et al. and Marconi & Tortora demonstrated the highest performance with an efficiency of

approximately 90% in a relatively short time (60 minutes) under sunlight [29], [30], indicating high effectiveness under more practical and energy-efficient conditions.

Table 5. Degradation of Methylene Blue

| Ref. | Photocatalyst Material (Best) | Initial Concentration | MB | Catalyst Dosage | Light Source | Degradation Efficiency | Time |
|------|--|--------------------------|----|------------------------|--------------------------|---|-------------|
| [29] | NT2 (TiO ₂ NPs - Microemulsion) | 2.5 × 10 ⁻⁵ M | | 0.6 g/L | Sunlight (Solar) | ~90% | 60 minutes |
| [31] | T/AOT-0.08 (TiO ₂ Nanoparticles) | 20 ppm | | 10 mg (optimal: 30 mg) | UV Lamp (2 × 15 W) | 76% (reaching 89% at a dose of 30 mg) | 300 minutes |
| [32] | 1% Fe- TiO ₂ (Iron-doped Titania) | 5 ppm | | 1.0 g/L | Visible Light (13 W LED) | 57%-80% (depending on irradiance intensity) | 180 minutes |
| [30] | NT2 (TiO ₂ NPs - Microemulsion) | 2.5 × 10 ⁻⁵ M | | 0.6 g/L | Sunlight (Solar) | 90% | 60 minutes |

This indicates that an efficient system can accelerate the degradation process without requiring extended reaction time. In contrast, the study by Honarmand et al. showed an initial efficiency of 76% that could increase to 89% with catalyst dose optimization, but required a longer time (300 minutes) and a UV light source, indicating a high energy dependency [31]. Despite the longer reaction time, the efficiency achieved is only comparable, suggesting that increasing time alone is not the dominant factor in improving degradation performance. Meanwhile, the study by Rosa et al. using Fe- TiO₂ showed varying efficiencies (57-80%) under visible light, with an intermediate reaction time (180 minutes), indicating an enhanced response to visible light, though not yet as effective as the microemulsion-based system [32]. These results indicate that reaction time contributes to degradation efficiency, but its effect is closely linked to the photocatalyst characteristics and irradiation conditions. The type of light source used has a significant impact on degradation efficiency because TiO₂ requires sufficient photon energy to excite electrons from the valence band to the conduction band, generating electron-hole pairs that initiate the formation of reactive species. Sunlight achieves better efficiency than other light sources because it provides a broader spectrum of irradiation, including both UV and visible light, as well as higher photon intensity, which enhances electron excitation and increases the generation of reactive oxygen species responsible for methylene blue degradation.

Discussion

The discussion of the results of this systematic literature review indicates that the microemulsion method plays a very significant role in the synthesis of TiO₂-based materials, particularly in controlling the physicochemical characteristics that directly influence photocatalytic performance. However, when considering the literature search strategy that also included Fe₃O₄, there appears to be a discrepancy between the initial search design and the selected articles. From the PRISMA selection process, only a small portion of the articles were truly relevant, and the majority still focused on TiO₂ without explicit integration with Fe₃O₄. This indicates that studies on the combination of the microemulsion method with the Fe₃O₄/TiO₂ system for methylene blue degradation remain limited, so the discussion predominantly centers on TiO₂ as the primary material. In general, the microemulsion approach works through the formation of micelles that function as nanoreactors, where the size and stability of the droplets determine the final particle size. The variety of microemulsion types used in the literature, such as water-in-oil (W/O) systems, conventional microemulsions, and microemulsion electrospinning, indicates that there is no fully standardized approach yet. However, all these methods share the same principle: utilizing a stable dispersion system to control particle growth with greater precision than conventional methods. The scarcity of literature directly examining Fe₃O₄ in these systems also indicates that research remains fragmented between studies on material synthesis and composite applications.

Microemulsion composition parameters such as surfactant type, concentration, phase ratio (Rv/Rw), and the presence of co-surfactants play a crucial role in determining system stability. The use of cationic surfactants such as CTAB and CPB, anionic surfactants such as AOT, and nonionic surfactants such as TX-100 demonstrates that surfactant selection is tailored to the chemical interaction requirements between the precursor and the reaction medium. Co-surfactants such as butanol and 1-pentanol serve to reduce interfacial tension, thereby increasing the flexibility of the surfactant layer and enabling the formation of more stable droplets. However, not all studies report these parameters in detail, particularly viscosity and emulsification energy. This constitutes a methodological limitation because these parameters significantly influence precursor diffusion, nucleation rates, and crystal growth in microemulsion systems, including the potential formation of composites such as Fe₃O₄/TiO₂.

In terms of material characteristics, the microemulsion method has proven capable of producing nanoparticles with a relatively homogeneous size distribution and controlled porosity. A study by Zhang et al. demonstrated that the microemulsion electrospinning approach can produce TiO₂ nanofiber structures with hierarchical pores and small particle sizes (17.8–20.1 nm), as well as a stable anatase phase at a calcination temperature of 500 °C [28]. This structure strongly supports photocatalytic processes by increasing surface area and reactant accessibility. Meanwhile, the W/O approach, as in Marconi et al., yields a high surface area despite aggregation, indicating that microstructural control is more important than macroaggregate size. Variations in synthesis conditions and resulting physicochemical properties, such as particle size, crystallinity, and surface area, indicate that these parameters are closely related to the effectiveness of photocatalytic performance. However, since few studies have integrated Fe₃O₄, the direct influence of magnetic properties on particle morphology and distribution has not yet been comprehensively analyzed.

The calcination temperature is a key factor in determining the crystal phase. In the range of 450 °C - 600 °C, a transformation from anatase to rutile occurs, which affects photocatalytic activity. Anatase is more reactive, whereas rutile is more stable. In the context of Fe₃O₄/TiO₂ composites, controlling this temperature becomes increasingly important as it has the potential to influence interphase interactions and composite stability, although this has not been extensively discussed in the analyzed literature. Modifications through metal doping, such as with Zr and Fe, indicate that performance improvements are not always linear. A study by Honarmand et al. showed that Zr doping enhances homogeneity without altering the main phase [31], whereas Rosa et al. demonstrated that Fe doping increases surface area but induces a mixed phase [32]. These differences suggest that structural and compositional modifications influence not only physical properties but also the resulting photocatalytic efficiency. This suggests that modification strategies remain primarily focused on TiO₂, rather than on integration with Fe₃O₄ as a functional composite.

At the application stage, the relationship between material characteristics and methylene blue degradation performance indicates that microemulsion-based materials exhibit high efficiency, reaching ~90% degradation within 60 minutes under sunlight, as reported by Marconi et al. and Marconi & Tortora. This result suggests that degradation efficiency increases with irradiation time, where pore structure and surface area act as dominant factors controlling adsorption and photocatalytic activity [29], [30]. However, comparison across different systems indicates that high degradation efficiency can be achieved within shorter reaction time when the photocatalyst is more effective, while longer reaction time does not necessarily result in significantly higher efficiency. Meanwhile, doped systems require more specific conditions and generally longer degradation times. Systems that require longer reaction times tend to achieve only comparable efficiency, indicating that photocatalyst characteristics and irradiation conditions have a more dominant influence than reaction time alone. However, without the presence of Fe₃O₄ in the analyzed system, aspects such as ease of magnetic separation and potential electron synergy have not been fully explored in this study.

Implications

The results indicate that the microemulsion method has proven effective in enhancing the characteristics of TiO₂ as a primary photocatalyst, particularly in producing nanoparticles, high porosity, and a large surface area, which contribute to the efficiency of methylene blue degradation. The recommended characteristics are small-sized anatase-phase TiO₂ (20–50 nm) with a mesoporous or hierarchical structure. Limitations in the literature regarding Fe₃O₄ indicate that the integration of Fe₃O₄ into microemulsion-based systems remains an area for further research. Therefore, future development should focus on exploring Fe₃O₄/TiO₂ combinations to investigate the synergistic effects between magnetic properties and photocatalytic activity, particularly regarding enhanced efficiency and ease of catalyst separation. Additionally, standardization of synthesis and testing parameters is required so that the influence of each component (TiO₂ and Fe₃O₄) can be analyzed more clearly. Future research will focus on integrating the microemulsion method with more controlled composite systems and testing under practical conditions, such as the use of sunlight and real wastewater.

CONCLUSION

The results indicate that the microemulsion method significantly influences the characteristics of TiO₂-based materials, particularly in terms of nanoparticle size, more homogeneous distribution, and the formation of high porosity and surface area. These characteristics were found to directly contribute to improved photocatalytic performance in the degradation of methylene blue, specifically through enhanced initial adsorption and reaction efficiency under irradiation. However, based on the results of the systematic review, most of the analyzed articles focused solely on TiO₂ without the integration of Fe₃O₄; thus, a direct relationship between the microemulsion method and the characteristics of Fe₃O₄/TiO₂ composites cannot yet be empirically proven from the available literature. Thus, the influence of the microemulsion method identified in this study more accurately represents the TiO₂ system as the primary material, particularly regarding the control of morphology, crystal phase (anatase–rutile), and surface area, which determine degradation performance. Conceptually, these results still indicate that if the microemulsion method is applied to Fe₃O₄/TiO₂ composite systems, performance improvements may occur through a combination of structural control of TiO₂ and the additional properties of Fe₃O₄, such as ease of separation and the potential for enhanced electron transfer. Therefore, there is a clear research gap: the lack of studies that directly examine the effect of the microemulsion method on Fe₃O₄/TiO₂ composites and its relationship to the degradation efficiency of methylene blue. Consequently, further research is needed to experimentally validate this potential.

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