

Surface Functionalization of Silica Materials for Enhanced Selectivity in Dye Adsorption. A Review of Chemical Modification Strategies

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Abstract

The discharge of synthetic dyes from industrial effluents represents a persistent environmental challenge because of their high toxicity, chemical stability, and resistance to conventional biodegradation. At low concentrations, these dyes can disrupt aquatic ecosystems, lower water quality, and threaten human health. Among available treatment methods, adsorption has emerged as an effective and versatile method for dye removal because it offers simple operation, low operating costs, and produces almost no secondary environmental contamination. Silica-based materials, such as mesoporous and nanoparticulate forms, are highly effective adsorbents due to their large surface area, adjustable porosity, and chemical stability. However, pristine silica has limited adsorption capacity and poor selectivity for specific dyes, limiting its practical use. To overcome these limitations, there have been extensive studies on the surface functionalization of silica, which optimizes surface chemistry and improves dye adsorption performance. Functionalization strategies such as organosilane grafting, polymer modification, metal and metal-oxide incorporation, and bio-based approaches employ mechanisms such as electrostatic attraction, hydrogen bonding, π - π interactions, and metal coordination. These modifications enhance adsorption efficiency and enable the selective removal of anionic, cationic, and non-ionic dyes. This review summarizes recent advances in chemical modification strategies for silica-based adsorbents, highlighting how different surface functional groups, structural characteristics, and dye selectivity correlate. It assesses the regeneration and reusability of functionalized silica materials and discusses current challenges and future directions. This review also integrates modern research insights to provide a comprehensive design framework for developing environmentally sustainable multifunctional silica adsorbents that selectively remove dyes from industrial wastewater.

Keywords: Silica functionalization; Dye adsorption; Selective removal; Surface modification strategies; Wastewater treatment

1 Introduction

Global industrial demand for synthetic dyes continues to rise in textile production, paper manufacturing, leather processing, plastic production, and the pharmaceutical industries. These dyes are widely used because of their bright colours, chemical stability, and resistance to degradation. However, the extensive use of these materials generates substantial volumes of coloured wastewater, which poses an environmental problem. Even at low concentrations, dyes can obstruct light penetration in aquatic ecosystems, disrupting photosynthesis, altering oxygen production, and negatively affecting aquatic plants and animals [1]. Several classes of synthetic dyes, such as azo, anthraquinone, and triarylmethane dyes, are known to cause toxic effects in humans, including mutagenicity, carcinogenicity, and endocrine disruption [2,3]. These environmental and health implications highlight the urgent need for effective wastewater treatment technologies for the selective removal of dyes.

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Among the various physicochemical and biological methods developed for dye removal, adsorption has emerged as the widely accepted method because of its easy operation, effective performance, and its safe impact on the environment [4]. The adsorption method has become the most popular choice for dye removal because of its easy operation, high effectiveness, and low environmental impact. Adsorption effectively removes dyes without producing secondary pollutants, making it an ideal solution for treating chemically stable dyes that resist conventional degradation methods [5]. Activated carbon, zeolites, clays, and metal oxides are some of the adsorbents that have been extensively explored. The unique physicochemical properties of silica-based materials have attracted growing interest because they possess high surface area, mechanical and thermal stability, tunable porosity, and easy modified surface chemistry for functionalization [6,7].

Pristine silica surfaces are dominated by silanol (Si-OH) groups, which can interact with dye molecules primarily through hydrogen bonding, van der Waals forces, and electrostatic interactions. However, these interactions become weak with non-selective behaviour, which restricts both adsorption capacity and specificity towards dye types [7]. For instance, unmodified mesoporous silica demonstrates low dye attraction towards cationic dyes in the presence of competing anions or anionic dyes under neutral pH conditions [6]. The solution to these restrictions requires surface functionalization strategies, which introduce specific chemical groups that improve the binding affinity, selectivity, and adsorption capability of silica-based adsorbents [8].

Surface functionalization of silica can be achieved through chemical grafting, co-condensation, post-synthetic modification, and polymer/ligand immobilization. These modifications enable the creation of multiple functions through which amino thiol, carboxyl, sulfonic, phosphonic, and quaternary ammonium groups can interact with cationic, anionic, and neutral dye molecules [9]. Amino-functionalized silica shows improved electrostatic attraction toward anionic dyes (methyl orange and Congo red). In contrast, sulfonic acid-modified silica shows a stronger tendency to adsorb cationic dyes (methylene blue) [10]. Hydrophobic dye adsorption improves with amphiphilic modifications via lipophilic interactions, thereby allowing the removal of more dye classes [11,12]. These improvements demonstrate how targeted surface chemistry is essential in attaining efficient and selective dye removal from complex wastewater.

Polyelectrolyte and polymer grafting have emerged as complementary strategies for improving selectivity and regeneration [13]. Silica particles with polycation coatings such as PDADMAC and polyaniline (PANI) show increased affinity for anionic dyes because of electrostatic attraction and hydrogen bonding, while maintaining high reusability across multiple adsorption-desorption cycles. Immobilizing biopolymers, such as chitosan, onto functionalized silica surfaces combines natural polymer selectivity with silica's high surface area and stability, resulting in improved adsorption performance for both cationic and anionic dyes [14,15]. Surface-specific aerogel fibres have been designed to provide continuous adsorption and degradation of dye pollutants, showing their potential for large-scale wastewater treatment applications [16,17].

Despite these advancements, challenges persist. Many functionalized silica adsorbents are developed under laboratory conditions using model dyes, while their performance in real industrial effluents containing complex mixtures of dyes, salts, surfactants, and organic contaminants is less explored [18]. Additionally, there is a lack of a complete understanding of how different surface modification densities, functional group distributions, and pore accessibility impact both adsorption kinetics and thermodynamic processes, resulting in varying adsorption efficiency across studies [19]. The large-scale synthesis of functionalized silica requires both economic and environmental impact assessments to determine its actual application viability. Addressing these gaps, research has focused on the combination of advanced surface chemistry with sustainable synthesis approaches. For example, silica derived from agricultural residue, such as rice husk, functionalized with amine groups for efficient MB dye adsorption, provides a cost-effective and sustainable alternative to conventional silica materials [20]. Microwave-assisted synthesis and green post-synthetic modifications have also been studied to improve access to functional groups while reducing energy use and chemical waste [21,22]. Collectively, these studies highlight a growing trend toward sustainable, selective silica-based adsorbents for the removal of industrial dyes.

This review examines recent developments in chemical surface functionalization of silica materials for dye adsorption. It identifies research trends, emerging strategies, and current gaps in the field by analyzing how functional groups and structural characteristics influence dye selectivity. The insights provided in this review are expected to guide future efforts toward designing highly efficient, selective, and sustainable silica-based adsorbents for industrial wastewater treatment.

2 Silica Materials and Surface Chemistry

Silica materials serve as effective adsorbents for dye removal due to their high surface area, adjustable porosity, chemical stability, and mechanical strength. The common forms of silica materials include amorphous silica, mesoporous silica (MCM-41 and SBA-15), nanoparticles, and bio-derived silica from agricultural waste products such as rice husk and sugarcane bagasse [23,24]. Their surface chemistry is primarily characterized by silanol ($\equiv\text{Si}-\text{OH}$) groups (figure 1), which determine hydrophilic properties, surface charge, and reactivity, enabling hydrogen bonding with dyes while providing bonding sites for covalent functionalization. Mesoporous silica, which has a high surface area ($700 - 1000 \text{ m}^2/\text{g}$), an organized pore structure, and adjustable pore size ($2 - 10 \text{ nm}$), enables rapid dye penetration and effective functional group loading, leading to selective adsorption [23,25]. However, at neutral and alkaline pH, unmodified silica surfaces are negatively charged, limiting interaction with anionic dyes, while cationic dyes are primarily adsorbed through electrostatic attraction. Chemical functionalization with amino, thiol, carboxyl, and polymeric groups improves selective dye binding and the total removal efficiency [26]. Bio-derived silica offers a sustainable, low-cost alternative, providing high surface area and reactive silanol groups for functionalization while valorizing agro-waste [27]. The relationship between silica type, surface chemistry, and structural properties must be understood to design adsorbents with high selectivity and performance in complex wastewater treatment systems.

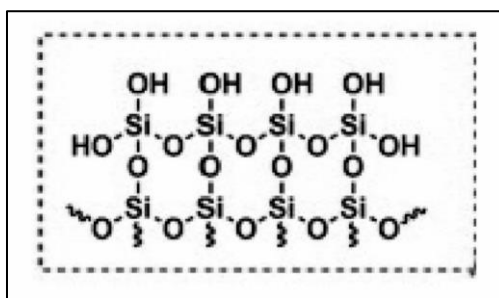


Figure 1 Silanol groups on silica fibre surface

3 Chemical Functionalization Strategies

Pristine silica has a high surface area and numerous silanol groups. However, its adsorption ability is often limited by weak interactions with dye molecules and the selectivity of specific dye types. To address these limitations, researchers have investigated various chemical functionalization methods to modify silica surface chemistry and improve dye-adsorbent interactions. Major functionalization strategies are discussed below.

3.1 Organosilane Functionalization

Organosilane grafting is a widely used method for modifying silica surfaces. Silane coupling agents such as 3-aminopropyltriethoxysilane (APTES), mercaptopropyltrimethoxysilane, and carboxyethylsilanes form stable $\text{Si}-\text{O}-\text{Si}$ bonds with surface silanol groups, covalently attaching functional groups that enable optimized interactions with specific dye molecules [28,29]. Amino-functionalized silica effectively removes anionic dyes such as Congo Red, Methyl Orange, and Acid Orange through electrostatic attraction, and research shows that its adsorption capacity exceeds that of unmodified silica by more than threefold [30,31]. Thiol and carboxyl modifications introduce additional binding mechanisms, such as hydrogen bonding and chelation, which benefit sulphur-containing azo dyes and metal-dye complexes [32]. Quaternary ammonium functionalization establishes a permanent positive charge, improving anionic dye adsorption across various pH range while achieving exceptional selectivity for reactive dyes in textile wastewater [33]. This strategy provides chemical stability, simple preparation, and adjustable functional properties, but excessive silane application can cause pore blockage, reducing access to internal sites. Optimal surface coverage is important to balance functional density and adsorption performance.

3.2 Polymer-Grafted Silica Surfaces

Polymer grafting improves dye adsorption by increasing both the density and heterogeneity of functional groups on silica surfaces through either "grafting-to" (connecting pre-formed polymers) or "grafting-from" (surface-initiated polymerization) methods. Polyethyleneimine (PEI) provides abundant amine groups for strong electrostatic attraction of anionic dyes [34]. Polyaniline (PANI) and other conductive polymers introduce $\pi-\pi$ interactions with aromatic dye, enhancing the selectivity for compounds such as methylene blue and rhodamine B [35,36]. Chitosan-functionalized silica uses electrostatic, hydrogen-bonding, and hydrophilic interactions to remove both cationic and anionic dyes

across a wide pH range [37]. Polymer grafting enhances mechanical and chemical stability, resulting in improved pH tolerance compared to silane-based modifications.

3.3 Metal and Metal-Oxide Functionalization

The incorporation of metal ions (Fe^{3+} , Cu^{2+} , Zr^{4+}) and metal oxides into silica creates new coordination sites for selective dye binding that contain nitrogen, sulphur, and hydroxyl groups, especially azo and reactive dyes. Metal-functionalized silica material forms chelation complexes, resulting in enhanced adsorption strength and selectivity, whereas magnetic silica composites enable easy separation and regeneration [38]. Mesoporous metal-functionalized silica maintains its high surface area and pore accessibility, enabling efficient and precise dye removal from complex wastewater [39]. Key challenges include preventing metal leaching and secondary contamination, demonstrating the requirement for stable immobilization and surface protection techniques.

3.4 Bio-Based and Green Functionalization Approaches

Bio-based functionalization uses renewable resources such as amino acids, polysaccharides, and plant-derived ligands to create environmentally friendly silica surface modifications. Rice-husk silica functionalized with an amino or carboxyl group is capable of selectively adsorbing methylene blue, Congo Red, and Neutral Red dyes [40]. Biopolymer coatings, including chitosan and alginate, improve hydrophilicity and multiple binding mechanisms. Green synthesis methods, such as microwave-assisted and solvent-free grafting, achieve maximum functional group exposure while reducing energy consumption and chemical waste production [41]. These strategies offer sustainable, low-cost solutions that achieve high adsorption performance with precise selectivity.

3.5 Mechanistic Insights and Selectivity

Surface functionalization methods are designed to enhance specific target-dye interactions. Amino and quaternary ammonium silica modifications enable the adsorption of anionic dyes through electrostatic attractions, while carboxyl and sulfonic acid surface modifications target cationic dyes. Polymer and aromatic-functionalized silica materials show enhanced dye binding with synergistic functional groups through hydrogen bonding and π - π interactions [42]. Metal coordination provides strong dye binding for nitrogen, sulphur, and hydroxyl-containing dyes. The introduction of amphiphilic or polymer-grafting methods establishes hydrophobic interactions, enhancing the removal of non-polar dyes. The selection of suitable functional groups and grafting strategies enables the development of silica adsorbents for high selectivity and capacity in multi-component dye applications.

4 Dye Selectivity and Interaction Mechanisms

Dye adsorption on functionalized silica occurs via several interaction mechanisms that determine selectivity and efficiency. Charged dyes exhibit electrostatic interactions as their primary mechanism, while amino and quaternary ammonium groups attract anionic dyes, and carboxyl groups and sulfonic acid groups target cationic dyes, with their performance influenced by pH, ionic strength, and functional group density [43]. Hydrogen bonds between polar surface groups ($-\text{OH}$, $-\text{NH}_2$, and $-\text{COOH}$) and matching dye moieties improve the adsorption of non-ionic or weakly charged dyes and work in combination with electrostatic interactions [44]. π - π interactions between aromatic functional groups on polymer and polyaniline surfaces, and aromatic dyes, enhance the adsorption of compounds such as methylene blue and rhodamine B [36,44]. Metal coordination on metal-functionalized silica provides strong selective binding for nitrogen-, sulphur-, or hydroxyl-containing dyes, which include azo and reactive dyes, while magnetic composites enable separation and reuse [45]. The introduction of hydrophobic interactions through alkyl or amphiphilic groups improves the effectiveness of non-polar and neutral dye removal. The combination of electrostatic, hydrogen-bonding, and π - π and metal coordination and hydrophobic interactions through multi-mechanistic surfaces demonstrates enhanced adsorption capacity, selectivity, and overall performance in mixed-dye and actual wastewater systems, showing the critical role of surface functionalization and engineering [44,45].

5 Regeneration and Reusability

The practical application of functionalized silica adsorbents depends on their ability to be regenerated and reused without significant loss of capacity or selectivity. Common regeneration techniques such as pH swings, salt solutions, organic solvents, and mild thermal treatment interfere with electrostatic, hydrogen-bonding, π - π , and coordination interactions to enable the release of adsorbed dyes [46]. Functionalized silica maintains 75-90% of its adsorption capacity over multiple testing cycles, with polymer-grafted materials showing improved mechanical and chemical stability in comparison with silane-only modifications [47]. Stability of functional groups, pore accessibility, strength of dye-surface interactions, and competing ions or contaminants in wastewater are key factors affecting reusability.

Industrial dye-removal operations need optimized regeneration conditions and suitable functionalization methods to achieve maximum adsorbent longevity and ensure affordable and environmentally friendly operations.

6 Challenges and Future Perspectives

Despite significant progress, the application of functionalized silica adsorbents for practical purposes faces several challenges. They include functional group leaching, multi-dye selectivity in wastewater systems, high synthesis costs, and secondary contamination from metals or chemicals [48,49]. Future research should develop multifunctional materials and hierarchical materials that integrate electrostatic, hydrogen-bonding, π - π , metal coordination, and hydrophobic interaction for enhanced selectivity and adsorption efficiency. Sustainable functionalization methods such as bio-based ligands, microwave-assisted grafting, and solvent-free modifications are essential for reducing both environmental damage and manufacturing expenses. The development of advanced regeneration methods, magnetic or easily separable composites, and pilot-scale experiments that test real wastewater systems is required to ensure stability and practical utilization. Computational design and machine learning can guide scientists in selecting appropriate functional groups and surface structures, which will help in the development of optimized adsorbents for treating complex industrial wastewater. The integration of these approaches is expected to advance functionalized silica toward sustainable, high-performance, and selective dye-targeted removal solutions.

7 Conclusion

Surface-functionalized silica materials present an effective solution for the selective removal of synthetic dyes from industrial wastewater because of their combination of high surface area, adjustable porosity, and chemical stability. Functionalization strategies such as organosilane grafting, polymer modification, metal/metal-oxide incorporation, and bio-based approaches optimize specific dye molecule interactions through hydrogen bonding, π - π interactions, electrostatic attraction, and metal coordination, which improve adsorption capacity and selectivity. Hierarchical and mesoporous structures enhance active-site accessibility, while polymer and bio-based modifications introduce alternative pathways to and pH tolerance. The adsorbents demonstrate regeneration and reusability ability under mild chemical or thermal treatments, although their long-term effectiveness relies on both functional group stability and wastewater composition. Key challenges that industrial operations face include synthesis scale-up, complex multi-dye system stability maintenance, leaching prevention, and performance optimization under actual conditions. Future research should prioritize creating multifunctional, environmentally sustainable silica adsorbents, coupled with advanced regeneration techniques, pilot-scale testing, and computational design to achieve practical, efficient dye removal. Functionalized silica provides an adaptable, high-performing solution to tackle environmental and public health challenges of synthetic dye contamination.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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