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From Pristine Silica to Multifunctional Adsorbents: A Systematic Review of SiO_2 Modification Strategies for Methyl Orange Removal

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Abstract

Silica (SiO_2)-based adsorbents have been increasingly developed for methyl orange (MO) removal owing to their chemical stability, controllable pore structures, and ease of surface modification. However, the relationships among modification strategies, surface characteristics, adsorption performance, and adsorbent–adsorbate interactions have not been comprehensively examined. This study aims to analyze the development of SiO_2-based adsorbents for MO removal through a systematic literature review (SLR). The literature search covered publications from 2011 to 2026 and yielded 19 articles for preliminary synthesis. The adsorbents were classified into six categories: pristine SiO_2, porous/mesoporous SiO_2, organic-functionalized SiO_2, metal/metal oxide–SiO_2 composites, magnetic SiO_2, and SiO_2–biopolymer hybrids. The review reveals a shift from exploiting physical material characteristics toward surface chemical engineering and multifunctional adsorbent design. Reported adsorption capacities ranged from 18.98 to 588 mg/g, although direct comparisons were limited by differences in experimental conditions and definitions of adsorption capacity. Pseudo-second-order kinetics and Langmuir isotherms were frequently reported, while the proposed adsorption mechanisms involved electrostatic interactions, hydrogen bonding, pore filling, and functional-group-specific interactions. Surface charge regulation through amine and quaternary ammonium functionalization emerged as an important approach to improving MO affinity. Nevertheless, limited testing in real wastewater, inconsistent mechanistic evidence, and insufficient long-term regeneration assessment remain significant challenges. Future research should prioritize selective functionalization, standardized performance assessment, and more sustainable synthesis to support practical wastewater treatment applications.

Keywords: silica-based adsorbents, methyl orange, wastewater treatment

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Introduction

The textile and textile products (TTP) industry makes a substantial contribution to the Indonesian economy. The Coordinating Ministry for Economic Affairs of the Republic of Indonesia (2025) reported that the sector employed approximately 3.75 million workers and generated USD 6.92 billion in export earnings (Pemerintah Dorong Penyusunan Grand Design Penguatan Industri Tekstil Dan Pakaian Jadi Berdaya Saing Global - Kementerian Koordinator Bidang Perekonomian Republik Indonesia, n.d.). However, these activities also make wastewater management a major concern, particularly during textile dyeing. Commonly used colorants include azo dyes containing azo chromophores. Methyl orange (MO) is an anionic azo dye frequently used as a model compound to evaluate wastewater treatment technologies. Although MO cannot be assumed to represent the entire composition of textile wastewater, its aromatic structure and sulfonate groups make it relevant for investigating interactions between anionic dyes and adsorbent surfaces (Liu et al., 2013a; Wang et al., 2021b).(*Pemerintah Dorong Penyusunan Grand Design Penguatan Industri Tekstil Dan Pakaian Jadi Berdaya Saing Global - Kementerian Koordinator Bidang Perekonomian Republik Indonesia*, n.d.)(Liu et al., 2013a; Wang et al., 2021b)

The presence of azo dyes at certain concentrations in water bodies can hinder sunlight penetration, disrupt photosynthesis in aquatic organisms, and affect ecosystem balance. Moreover, the aromatic structure and azo bond of MO raise toxicological concerns, including the potential formation of transformation products during degradation. Various dye wastewater treatment methods have been investigated, including coagulation–flocculation, membrane filtration, biological treatment, advanced oxidation, photocatalysis, and adsorption. Akter and Islam (2022) reported that electrocoagulation using iron electrodes with added Fe^{2+} achieved 100% MO removal under specific experimental conditions (Akter & Islam, 2022). Ding et al. (2017) developed a nanofiltration membrane incorporating a SiO_2 rejection layer that achieved 97.6% MO rejection (Ding et al., 2017). Biological treatment also shows potential: Haque et al. (2024) identified biofilm-forming bacteria capable of degrading and detoxifying MO through azo-bond cleavage (Haque et al., 2024). Fenton oxidation reportedly removed more than 97.8% of MO within 15 minutes at pH 2.79, whereas an Ag–TiO_2 photocatalyst achieved 100% decolorization within 70 minutes and 75.43% mineralization within 90 minutes (W. Zhang et al., 2024). These findings indicate that MO can be removed through separation or degradation mechanisms; however, treatment performance must also be evaluated in terms of operating conditions, energy and chemical requirements, residual waste generation, and treated-water quality.(Akter & Islam, 2022)(Ding et al., 2017)(Haque et al., 2024)(W. Zhang et al., 2024)

Among these technologies, adsorption is considered attractive because of its operational simplicity, flexibility, and relatively straightforward separation of dyes from the aqueous phase (Bilal et al., 2022; Crini & Lichtfouse, 2018; Yagub et al., 2014). Silica (SiO_2) is a promising adsorbent owing to its chemical stability, diverse raw material sources, and the availability of synthesis methods capable of producing porous structures with high surface areas (Haider et al., 2022). In addition to commercial precursors, SiO_2 can be obtained from silica-rich biomass waste. Haider et al. (2022) demonstrated that rice husk ash could serve as a silica source for dye adsorption, while Dokkathin et al. (2025) synthesized SiO_2 nanoparticles from sugarcane bagasse ash and modified them with CTAB to enhance affinity toward anionic dyes (Dokkathin et al., 2025). Such alternative feedstocks may add value to agricultural residues and reduce reliance on commercial silica precursors, although overall economic feasibility still depends on energy consumption, chemical inputs, and purification steps. Zhang et al. (2023) reported an MO

adsorption capacity of 69.40 mg/g for SiO_2 nanoparticles (X. Zhang et al., 2023). Wang et al. (2021) further demonstrated that functionalizing silica with a multi-hydroxyl-containing gemini surfactant substantially improved adsorption capacity (Wang et al., 2021c). Accordingly, SiO_2-based adsorbent development should seek to improve MO affinity while maintaining ease of production and reusability.(Bilal et al., 2022; Crini & Lichtfouse, 2018; Yagub et al., 2014)(Haider et al., 2022)(Haider et al., 2022)(Dokkathin et al., 2025)(X. Zhang et al., 2023)(Wang et al., 2021c)

Research developments indicate that SiO_2 modification can substantially change MO adsorption performance. Liu et al. (2013) modified SiO_2 nanoparticles with quaternary ammonium polyethylenimine (QPEI), obtaining a maximum adsorption capacity of 105.4 mg/g and an equilibrium time of approximately 10 minutes (Liu et al., 2013b). Wang et al. (2021) subsequently extended organic functionalization using a multi-hydroxyl-containing gemini surfactant, increasing MO adsorption capacity from 64.72 to 401.88 mg/g (Wang et al., 2021c). Subsequent advances have involved not only the introduction of organic functional groups but also the development of multifunctional composites. Li et al. (2024) developed magnetic Fe_3O_4@SiO_2, which enables adsorbent separation using an external magnetic field, and reported an adsorption capacity of 182.503 mg/g under the study's optimum conditions (Li et al., 2024a). In the same year, Taweekarn et al. (2024) incorporated mesoporous silica nanoparticles into a starch cryogel matrix and reported approximately 99% MO removal in synthetic solutions and 93% in real wastewater samples (Taweekarn et al., 2024). Dokkathin et al. (2025) later showed that modifying bagasse-derived silica with cetyltrimethylammonium bromide (CTAB) changed the surface zeta potential from −28.96 to +12.06 mV (Dokkathin et al., 2025). These findings demonstrate that SiO_2 modification concerns not only surface area enhancement but also charge regulation, active-site formation, pore accessibility, and ease of material separation. Nevertheless, differences in experimental conditions prevent adsorption capacities from being directly compared as absolute indicators of material superiority.(Liu et al., 2013b)(Wang et al., 2021c)(Li et al., 2024a)(Taweekarn et al., 2024)(Dokkathin et al., 2025)

Overall, the development of SiO_2-based adsorbents reflects a shift from exploiting textural properties toward controlling surface chemistry and designing multifunctional composites. Mesoporous structures provide mass-transfer pathways for MO molecules, whereas amine or quaternary ammonium functionalization can strengthen electrostatic interactions with MO sulfonate groups. Incorporating metal oxides, magnetic materials, and biopolymers can further expand the range of active sites while introducing additional functions, including easier separation and improved operational stability. Future modification strategies may therefore focus on more selective surface charge control, complementary functional groups, hierarchical pore structures, and more sustainable silica sources. However, improvements in adsorption capacity are not always accompanied by sufficient evidence to distinguish the contributions of electrostatic interactions, hydrogen bonding, hydrophobic interactions, and intraparticle diffusion. This systematic literature review therefore examines the development of SiO_2-based adsorbent modifications for MO removal, emphasizing the relationships among modification strategies, surface-property changes, adsorption performance, and adsorbent–adsorbate interaction mechanisms. The synthesis is expected to provide a scientific basis for developing more selective, effective, and sustainable SiO_2 adsorbents.

Research Methods

Review design. This review followed the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA 2020) guidelines developed by Page et al. (2021) to support systematic

and transparent literature identification, selection, assessment, and reporting (Page, McKenzie, et al., 2021; Page, Moher, et al., 2021). The review focused on four aspects: the temporal development of SiO_2 modification strategies; relationships between adsorbent physicochemical characteristics and MO adsorption performance; interactions between MO molecules and adsorbent active sites; and research gaps that could inform future material development. Unlike reviews that primarily compare adsorption capacities, this study emphasizes the links among modification type, changes in surface properties, adsorption behavior, and the strength of experimental evidence supporting proposed mechanisms.(Page, McKenzie, et al., 2021; Page, Moher, et al., 2021)

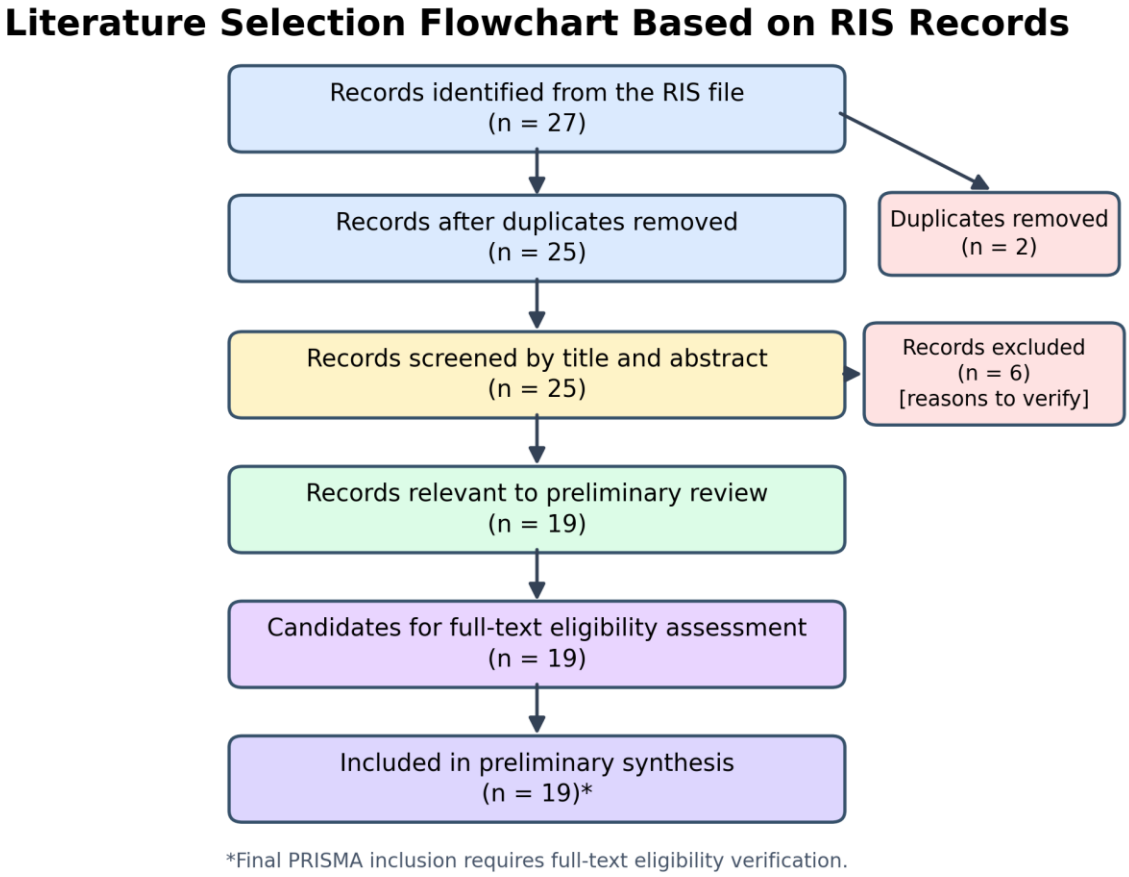

Figure 1. Flowchart of literature identification and preliminary screening for the systematic review of SiO_2-based adsorbents for methyl orange removal.

Literature selection. The literature search was designed to cover Scopus and Web of Science, supplemented by searches of ScienceDirect, SpringerLink, and ACS Publications. Publications dated January 2011 to October 2026 were considered. Search terms included “methyl orange”, “acid orange 52”, “silica”, “silicon dioxide”, “SiO_2”, “mesoporous silica”, “silica nanoparticles”, and “adsorption”, combined using the Boolean operators AND and OR. Both pristine and modified SiO_2 were considered to identify developments in modification strategies and their effects on MO adsorption performance.

Articles were selected according to PRISMA 2020 through identification, duplicate removal, title and abstract screening, and full-text eligibility assessment. Inclusion criteria comprised peer-reviewed primary research articles using SiO_2-based adsorbents and reporting experimental MO adsorption data from aqueous solutions. Studies reporting only photocatalytic degradation or other treatment processes without separately identifiable adsorption data, review articles, and duplicate publications were excluded. Selection stages and reasons for exclusion were documented in the PRISMA 2020 flow diagram to promote transparency and reproducibility.

Data extraction and synthesis. Data were systematically extracted from the selected studies, including publication year, SiO_2 modification type, adsorbent physicochemical characteristics (BET surface area, pore structure, functional groups, and surface charge), experimental conditions, and MO adsorption performance and mechanisms. Study quality was assessed in terms of the completeness of material characterization, clarity of experimental conditions, and evidence supporting the proposed adsorption mechanisms. The data were then synthesized narratively and comparatively to examine the development of SiO_2 modification strategies and the relationships among surface properties, adsorption capacity, and adsorbent–adsorbate interactions. Differences in experimental conditions were considered when interpreting the findings. This synthesis was used to identify research gaps and propose directions for developing more effective, selective, and sustainable SiO_2-based adsorbents.

Bibliographic verification. Selected bibliographic fields were checked against DOI or publisher records when the RIS entry contained obvious formatting inconsistencies. In particular, the DOI for the pyrazole amide-functionalized silica paper was normalized to 10.1002/aoc.70254, and the Armid et al. study is cited as 2022 based on the journal metadata associated with DOI 10.29303/aca.v6i2.159. These corrections affect citation formatting rather than the substantive findings extracted from the supplied records.

Results and Discussion

Evolution of SiO_2-Based Adsorbent Modifications for Methyl Orange Adsorption

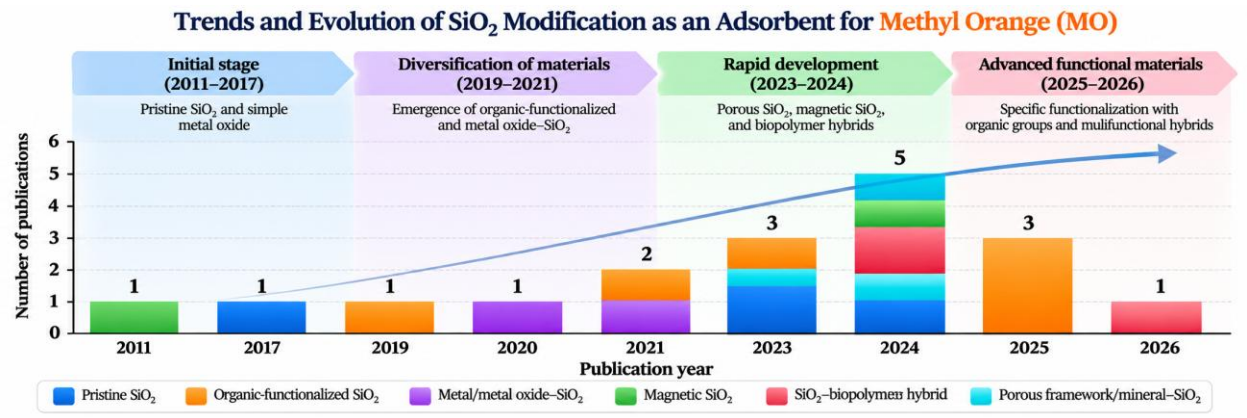

Figure 2. Temporal distribution and evolution of SiO_2-based adsorbent modification strategies for methyl orange removal during 2011–2026

Figure 2 illustrates developments in SiO_2-based adsorbent modifications for MO adsorption from 2011 to 2026. During 2011–2021, research remained relatively limited, with one or two publications in each represented year. Publication numbers began to increase in 2023 and reached their highest level in 2024. Three publications were recorded in 2025 and one in 2026. The increase in both publication volume and material diversity, particularly during 2023–2024, suggests growing research interest in engineering SiO_2-based materials to enhance MO adsorption performance.

As shown in Figure 2, the research trajectory can be divided into four main stages. The initial stage (2011–2017) was characterized by pristine SiO_2 and relatively simple metal oxide composites. At this stage, physical properties such as specific surface area, pore structure, and the presence of silanol (Si–OH) groups were important to adsorption. However, silica surfaces tend to be negatively charged at pH values above the point of zero charge, which can limit electrostatic attraction toward anionic MO. This limitation encouraged the development of modification strategies to improve surface affinity for MO molecules.

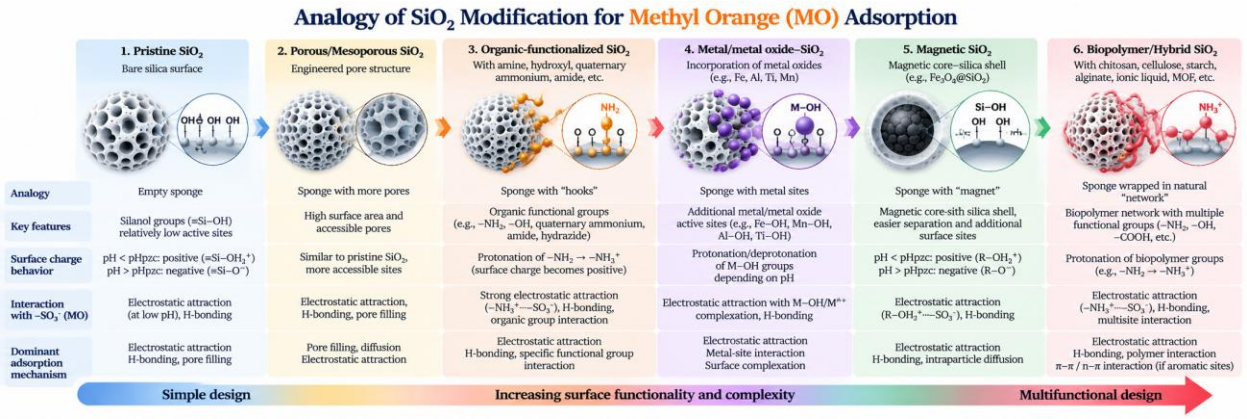

Figure 3. Schematic Illustration of SiO_2 Modification Strategies and Their Proposed Adsorption Mechanisms for Methyl Orange Removal

As illustrated in Figure 3, the material diversification stage (2019–2021) involved the increasing use of organic functionalization and SiO_2 composites containing metals or metal oxides. These modifications aimed to introduce new functional groups and alter surface chemistry to promote more effective interactions between adsorbents and MO. Wang et al. (2021) reported that functionalizing silica nanoparticles with a multi-hydroxyl-containing gemini surfactant increased MO adsorption capacity from 64.72 to 401.88 mg/g (Wang et al., 2021c). This finding indicates that surface chemical engineering can substantially improve adsorption performance, not only by modifying pore characteristics but also by introducing additional interaction sites for MO molecules.(Wang et al., 2021c)

More rapid development occurred during 2023–2024, when modification strategies diversified to include porous SiO_2, magnetic composites, and biopolymer-based hybrids. These approaches aimed not only to increase adsorption capacity but also to improve operational properties, including separation and reuse. Li et al. (2024) developed magnetic Fe_3O_4@SiO_2 with an MO adsorption capacity of 182.503 mg/g (Li et al., 2024b), while Taweekarn et al. (2024) reported that mesoporous silica incorporated into starch cryogel achieved approximately 99% MO removal in synthetic solutions and 92.77% in real wastewater (Taweekarn et al., 2024). These findings suggest

a shift toward integrating textural properties, surface chemistry, and additional functionalities to support practical application.(Li et al., 2024b)(Taweekarn et al., 2024)

During 2025–2026, research increasingly explored more specific organic functionalization and multifunctional hybrid materials. These strategies focused on controlling surface properties by introducing active groups that could enhance MO affinity. For example, Dokkathin et al. (2025) reported that modifying bagasse ash-derived SiO_2 with cetyltrimethylammonium bromide (CTAB) changed the zeta potential from −28.96 to +12.06 mV (Dokkathin et al., 2025). This shift indicates a more positively charged surface, potentially strengthening electrostatic attraction toward the sulfonate ($–SO_3^-$) group of MO. The use of agricultural waste-derived silica also offers opportunities for more sustainable adsorbents, although chemical consumption, material stability, and regeneration performance require further evaluation.(Dokkathin et al., 2025)

Overall, the evolution of SiO_2 modification reflects a transition from relying on physical material characteristics to engineering surface chemistry and developing multifunctional composites. This transition indicates that MO adsorption effectiveness depends not only on surface area and porosity but also on functional groups, surface charge, active-site accessibility, and adsorbent stability. Future studies should therefore develop adsorbents with greater affinity and selectivity toward anionic dyes, good regeneration performance, and stable operation in real wastewater. Stronger mechanistic evidence from FTIR characterization, zeta potential or pH_pzc measurements, and pH-dependent adsorption studies is also needed to clarify the contributions of electrostatic attraction, hydrogen bonding, and diffusion to MO removal.

Table 1. Characteristics of the included studies

Study	Silica-based material	Reported performance	Kinetic model	Isotherm model	Key mechanism / feature
Li et al. (2024)	Fe3O4@SiO2 magnetic core–shell	182.503 mg g−1	Pseudo-second-order	Langmuir	Electrostatic attraction; hydrogen bonding; 83% capacity after 5 cycles
Mayangsari et al. (2026)	Fe3O4@SiO2@chitosan Schiff base; bagasse-derived SiO2	Not stated in abstract	Pseudo-second-order	Elovich	Electrostatic, H-bonding, π–π and n–π interactions; RSM optimization
Zhang et al. (2023)	Mesoporous SiO2 nanoparticles	69.40 mg g−1	Not stated	Langmuir, Freundlich, Redlich–Peterson, Temkin	pH, concentration, time and temperature effects; toxicity assessment
Armid et al. (2022)	Magnetic material–SiO2–zeolite composite	Not stated in abstract	Pseudo-second-order	Langmuir	Magnetic recovery; spontaneous, exothermic adsorption
Mohammadi & Sabourmoghaddam (2020)	TiO2–SiO2/alginate nanocomposite	68.85 mg g−1	Not stated	Langmuir	High surface area; spontaneous and exothermic adsorption
Ambreen et al. (2024)	Mn2O3/SiO2 nanocatalyst	Up to 99% MO degradation	Pseudo-first-order	Not applicable	Visible-light photocatalysis; pH-dependent performance
Wang et al. (2021)	Gemini-surfactant-functionalized organo-silica nanoparticles	401.88 mg g−1	Pseudo-second-order	Langmuir	Partitioning, electrostatic and OH−–π interactions
Dokkathin et al. (2025)	CTAB-modified silica nanoparticles from sugarcane bagasse ash	135.81 mg g−1	Not stated	Not stated	Surface charge reversal; selective anionic-dye adsorption
Taweekarn et al. (2024)	Mesoporous SiO2 in starch cryogel	18.98 mg g−1; 99% removal in 10 mg L−1 synthetic water	Pseudo-second-order	Langmuir	Electrostatic, hydrophobic and hydrogen-bonding interactions; tablet form
El Foulani et al. (2025a)	Hydrazide-functionalized silica	133 mg g−1	Pseudo-first-order	Not stated	Strong surface interaction; molecular simulation; 5-cycle regeneration

Study	Silica-based material	Reported performance	Kinetic model	Isotherm model	Key mechanism / feature
El Foulani et al. (2025b)	Pyrazole amide-functionalized silica	133.12 mg g−1	Pseudo-first-order	Redlich–Peterson	Acidic-surface interaction; molecular dynamics; >95% efficiency after 5 cycles
Rahmatpour et al. (2024)	Chitosan/xanthan gum–SiO2 hydrogel film	294 mg g−1	Pseudo-second-order	Langmuir	Electrostatic attraction and hydrogen bonding; 5 reuses
Salama (2023)	Amino-containing cellulose/silica microspheres	588 mg g−1	Pseudo-second-order	Langmuir	Amino-functional hybrid; sustainable cellulose–silica architecture
Deligeer et al. (2011)	Mesoporous γ-Fe2O3/SiO2 nanocomposite	476 mg g−1	Not stated	Langmuir and Freundlich	Mesoporosity; magnetic ferrimagnetic composite; rapid uptake
Arshadi et al. (2013)	Ferrocenecarboxaldehyde-functionalized SiO2–Al2O3	Not stated in abstract	Pseudo-second-order	Freundlich, Langmuir, Langmuir–Freundlich	Surface Fe3+ sites; spontaneous and endothermic adsorption
Otalvaro et al. (2019)	APTES-functionalized MCM-41 silica	Not stated in abstract	Not stated	Not stated	Protonated amines reverse surface charge; ionic-strength and stability effects
Sun et al. (2020)	SiO2@ZIF-67/CNTs composite	324 mg g−1 after calcination; 112 mg g−1 at 5 °C before calcination	First-order	Langmuir and Freundlich	Electrostatic interaction; hydrophobicity; ethanol regeneration
Alsaab et al. (2025)	Biogenic fibrous silica functionalized with crown-ether ionic liquid	507.1 mg g−1 for MO	Not stated	Langmuir	Sorghum-waste silica; dual adsorption; spontaneous, exothermic MO uptake
Shafqat et al. (2024)	L-proline- and furanacrylic-acid-grafted amino-silica	85.05% and 81.70% removal at pH 2	Pseudo-second-order	Freundlich	Acidic adsorption; organic grafting increases surface functionality

** Performance values are reported under the conditions stated in the source records and should not be interpreted as directly comparable because experimental conditions differed.*

Adsorption capacity: useful indicator, but not a stand-alone metric

According to Table 1, the reported MO adsorption capacities of SiO_2-based adsorbents vary considerably, ranging from 18.98 to 588 mg/g. Salama (2023) reported the highest capacity using cellulose/silica microspheres (Salama, 2023). The second-highest value, 507.1 mg/g, was reported by Alsaab et al. (2025) for biogenic fibrous silica functionalized with a crown ether ionic liquid (Alsaab et al., 2025). Deligeer et al. (2011) reported 476 mg/g for a mesoporous γ-Fe_2O_3/SiO_2 composite (Deligeer et al., 2011), while Wang et al. (2021) reported 401.88 mg/g for gemini surfactant-functionalized silica (Wang et al., 2021a). Conversely, Taweekarn et al. (2024) reported a capacity of 18.98 mg/g for a mesoporous silica–starch cryogel composite, although it achieved 99% MO removal in a synthetic solution (Taweekarn et al., 2024). These differences demonstrate that a high adsorption capacity does not necessarily correspond to high removal efficiency under specific application conditions. Adsorbent performance should therefore be assessed in relation to initial MO concentration, adsorbent dosage, pH, contact time, and solution matrix. Furthermore, experimental equilibrium capacity (q_e) must be distinguished from the maximum capacity estimated by isotherm modeling (q_{max}) to avoid misleading cross-study comparisons. Overall, the synthesis indicates that adsorption performance depends not only on SiO_2 porosity but also on functional groups and the compatibility of surface chemistry with MO molecular properties.(Salama, 2023)(Alsaab et al., 2025)(Deligeer et al., 2011)(Wang et al., 2021a)(Taweekarn et al., 2024)

Kinetic and isotherm behavior

Table 1 shows that the pseudo-second-order (PSO) model is the most frequently reported kinetic model for MO adsorption onto SiO_2-based adsorbents. Materials described by PSO kinetics

include magnetic Fe_3O_4@SiO_2 (Li et al., 2024b), gemini surfactant-functionalized silica (Wang et al., 2021a), cellulose/silica microspheres (Salama, 2023), and chitosan/xanthan gum–SiO_2 hydrogels (Rahmatpour & Hesarsorkh, 2024). Pseudo-first-order (PFO) kinetics were reported for hydrazide- and pyrazole amide-functionalized silica (El Foulani, Tarhouchi, Chafi, et al., 2025). Sun et al. (2020) also reported first-order kinetics for SiO_2@ZIF-67/CNTs (Sun et al., 2020). These differences indicate that kinetic behavior depends on surface characteristics, functional groups, and adsorption conditions. Although PSO kinetics are often associated with chemisorption, a good kinetic fit alone does not establish the formation of chemical bonds between the adsorbent and MO.(Li et al., 2024b)(Wang et al., 2021a)(Salama, 2023)(Rahmatpour & Hesarsorkh, 2024)(El Foulani, Tarhouchi, Chafi, et al., 2025)(Sun et al., 2020)

Langmuir was the most frequently reported isotherm model, while Freundlich, Redlich–Peterson, Temkin, and Elovich models were also identified for certain materials. Systems reported to follow Langmuir behavior included Fe_3O_4@SiO_2 (Li et al., 2024b; Mayangsari et al., 2026), TiO_2–SiO_2/alginate (Mohammadi & Sabourmoghaddam, 2020), cellulose/silica (Salama, 2023), γ-Fe_2O_3/SiO_2 (Deligeer et al., 2011), and several functionalized silica adsorbents. The Langmuir model suggests that a monolayer adsorption approximation involving assumed homogeneous sites can describe equilibrium data for some systems. However, the applicability of alternative models suggests possible heterogeneity in adsorption energies and surface interactions, particularly when SiO_2 is functionalized with organic groups or combined with other materials. Kinetic and isotherm models should therefore be understood as representations of adsorption behavior under specific experimental conditions rather than definitive evidence of molecular mechanisms.(Li et al., 2024b; Mayangsari et al., 2026)(Mohammadi & Sabourmoghaddam, 2020)(Salama, 2023)(Deligeer et al., 2011).

Role of pH and surface charge

Solution pH strongly influences MO adsorption because it controls the protonation of functional groups and the surface charge of adsorbents. As an anionic dye containing sulfonate groups ($–SO_3^-$), MO generally exhibits stronger electrostatic affinity for positively charged surfaces. Li et al. (2024) reported optimum adsorption at pH 2 using Fe_3O_4@SiO_2 (Li et al., 2024b), while Shafqat et al. (2024) obtained removal efficiencies of 85.05% and 81.70% at pH 2 using silica modified with L-proline and furanacrylic acid, respectively (Shafqat et al., 2024). Otalvaro et al. (2019) also demonstrated the importance of surface charge by introducing protonatable amine groups through APTES functionalization (Otalvaro et al., 2019). Dokkathin et al. (2025) reported that CTAB modification changed the zeta potential of SiO_2 from −28.96 to +12.06 mV, indicating a change in surface charge that could enhance interactions with anionic dyes (Dokkathin et al., 2025). These results highlight surface charge engineering as an important strategy for improving MO affinity. However, strongly acidic operating conditions may hinder practical wastewater treatment because they require pH adjustment and additional chemical consumption. Developing adsorbents that remain effective at pH values close to those of actual wastewater is therefore an important research priority.(Li et al., 2024b)(Shafqat et al., 2024)(Otalvaro et al., 2019)(Dokkathin et al., 2025).

Mechanistic interpretation

MO adsorption onto SiO_2-based materials generally involves a combination of electrostatic interactions, hydrogen bonding, hydrophobic interactions, and interactions involving π-electron systems. Electrostatic attraction is particularly relevant when SiO_2 surfaces are modified with positively charged or protonatable groups, increasing affinity for MO sulfonate groups. Hydroxyl,

amine, and other organic functional groups can also provide additional interactions between the adsorbent and adsorbate. Wang et al. (2021) identified partitioning, electrostatic interactions, and OH–π interactions in gemini surfactant-functionalized silica, while Mayangsari et al. (2026) proposed electrostatic interactions, hydrogen bonding, π–π interactions, and n–π interactions for Schiff base-functionalized Fe_3O_4@SiO_2–chitosan composites. El Foulani et al. (2025a, 2025b) further used molecular simulations to examine MO configurations and interactions with functionalized silica surfaces (El Foulani, Tarhouchi, Chafi, et al., 2025; El Foulani, Tarhouchi, Hammoudan, et al., 2025). Together, these studies suggest that SiO_2 modification can improve adsorption through several simultaneous interactions. Nevertheless, mechanistic interpretations should be supported by complementary characterization before and after adsorption, including FTIR, zeta potential, pH_pzc, and structural and porosity analyses. Kinetic or isotherm model fitting alone cannot confirm a particular adsorption mechanism. The stability of functional groups during regeneration is also important because changes in surface chemistry may reduce active-site availability and adsorption performance over repeated cycles.(El Foulani, Tarhouchi, Chafi, et al., 2025; El Foulani, Tarhouchi, Hammoudan, et al., 2025)

Sustainability and practical applicability

The development of SiO_2-based adsorbents increasingly emphasizes renewable feedstocks, ease of separation, and regeneration. Silica obtained from sugarcane bagasse ash (Dokkathin et al., 2025) and sorghum waste (Alsaab et al., 2025), together with cellulose, starch, chitosan, and xanthan gum matrices, illustrates the potential of alternative raw materials (Alsaab et al., 2025; Dokkathin et al., 2025). However, waste-derived or biopolymer-based feedstocks do not automatically ensure sustainability, as material synthesis and modification may still require substantial chemicals, solvents, and energy. From an operational perspective, Li et al. (2024) reported that Fe_3O_4@SiO_2 retained approximately 83% of its initial adsorption capacity after five cycles (Li et al., 2024b), while El Foulani et al. (2025b) reported more than 95% efficiency after five cycles for pyrazole amide-functionalized silica (El Foulani, Tarhouchi, Chafi, et al., 2025). Magnetic composites offer the additional benefit of easier adsorbent separation, while Taweekarn et al. (2024) demonstrated practical potential through tests on real wastewater, achieving 92.77% MO removal (Taweekarn et al., 2024). Nevertheless, most studies were conducted using controlled synthetic solutions, which may not adequately represent the effects of competing ions, dissolved organic matter, and variable wastewater composition. Future research should therefore prioritize real wastewater testing, long-term regeneration, functional-group stability, and assessments of chemical and energy consumption. Such efforts are needed to ensure that SiO_2-based adsorbents offer not only high laboratory adsorption capacities but also technical and environmental feasibility for wastewater treatment.(Alsaab et al., 2025; Dokkathin et al., 2025)(Li et al., 2024b)(El Foulani, Tarhouchi, Chafi, et al., 2025)(Taweekarn et al., 2024)

Research gaps and future directions

Five gaps emerge from the synthesis. First, experimental conditions are highly variable, making direct comparison of qmax values unreliable. A standardized reporting framework should include pH, ionic strength, initial concentration, adsorbent dose, temperature, equilibrium time, particle size, surface area, and regeneration protocol. Second, many studies rely on synthetic single-solute solutions. Competitive adsorption and real wastewater should be incorporated more routinely.

Third, the relationship between surface area and chemical functionality needs to be separated more carefully. Functionalization can increase affinity while simultaneously reducing pore volume or accessible surface area, as shown in amine-functionalized mesoporous silica. Fourth, regeneration studies should move beyond five cycles and quantify structural and chemical stability after repeated use. Finally, sustainability claims should be supported by process-level analysis, including precursor source, energy demand, chemical consumption, and waste generation. A future review could combine these standardized data with life-cycle assessment and techno-economic analysis to determine whether high-capacity silica hybrids remain advantageous when prepared at larger scale.

Conclusions

The reviewed literature shows that silica-based materials remain versatile platforms for methyl orange removal because their pore structure, surface charge, and functional groups can be deliberately modified. The main development has been from conventional mesoporous silica toward magnetic, organically functionalized, and biomass-derived hybrid materials. High adsorption capacities are generally associated with additional binding functionality rather than silica surface area alone. Langmuir and pseudo-second-order models are common, but the diversity of reported models confirms that adsorption behavior depends strongly on material chemistry and operating conditions. Future work should emphasize realistic wastewater matrices, standardized reporting, long-term regeneration, and sustainability assessment before high-performing laboratory materials are advanced toward practical treatment systems.

Author Contributions

IFU conceived the review, developed the review framework, screened and synthesized the literature, prepared the figures and tables, and wrote the manuscript. The author reviewed and approved the final version of the manuscript.

Conflict of Interest

The author declares no conflict of interest.

Declaration of Artificial Intelligence (AI)

An AI language tool was used during manuscript preparation to assist with language refinement, organization, and formatting. The author independently reviewed the literature, checked the extracted information against the supplied records and selected publisher/DOI metadata, revised the manuscript, and takes full responsibility for the accuracy, interpretation, and originality of the final content.

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