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APPLICATION OF GEOPOLYMERS AS EMERGING MATERIALS IN ADSORPTION TOWARDS ORGANIC DYES: A REVIEW

Organic dyes, widely used in industries such as textiles, are major pollutants in wastewater due to their non-biodegradable nature. They threaten human health and ecosystems, making their effective removal from effluents essential through advanced treatment methods. Indeed, adsorption has garnered significant attention for its high efficiency, simplicity, cost-effectiveness, and the reusability of adsorbents. Among various adsorbents, geopolymers with a three-dimensional aluminosilicate structure have gained attention as effective adsorbents for organic dyes removal, owing to their high efficiency, cost-effectiveness, and ability to be synthesized from industrial by-products. However, research on the operational factors affecting the adsorption performance of geopolymers remains limited. Addressing this gap will strengthen their application in advanced and eco-friendly wastewater treatment strategies. This paper reviews the synthesis and application of geopolymers materials for the adsorption of organic dyes. The key operational parameters, such as pH, adsorbent dosage, temperature, and initial pollutant concentration, are briefly reviewed to emphasize their critical impact on adsorption efficiency. Furthermore, the adsorption mechanisms responsible for organic dye removal are also being explained. These insights support the reported performance of geopolymers, which is comparable to other adsorbents. Hence, this supports the development of geopolymers as a promising reference for efficient wastewater treatment.

Keywords: Organic dyes; adsorption; wastewater

1. Introduction

Rapid industrialization and urban growth have greatly compromised water quality, primarily due to the release of substantial wastewater volumes and the influx of diverse pollutants into aquatic ecosystems. The need for freshwater is expected to surge by up to 55% from 2000 to 2050, fueled by the continued expansion of industrial activities [1]. This issue is concerning as it leads to environmental pollution. Wastewater contains numerous hazardous substances, including organic dyes, heavy metals, pathogenic bacteria, pharmaceuticals, pesticides, and other aromatic compounds [2,3]. Organic dyes, commonly used in industries such as printing, textiles, leather, and paper, contribute to wastewater with high levels of organic and dye content [4]. These activities have significantly contributed to the increased presence of dyes in the ecosystems. Wastewater from the textile industry, laden with harmful dyes originating from both textile manufacturing and dyestuff production, poses a significant environmental challenge. About 280,000 tonnes of dyes per year are being dumped into the environment, thus accelerating the problem of water pollution [5,6]. The aromatic chemical structures of these dyes contribute to their carcinogenic and mutagenic properties, presenting significant hazards to both human health and aquatic ecosystems [7-9]. Consequently, eliminating organic dyes from wastewater is essential before discharge to safeguard public health and the environment.

Several conventional methods are utilised to eliminate pollutants from wastewater, including coagulation, flocculation, precipitation, ozonation, filtration, and microbial treatments. Moreover, various separation techniques, such as adsorption, ion exchange, electrochemical processes, membrane bioreactors, reverse osmosis, advanced oxidation processes, and hybrid approaches, are widely implemented to improve treatment efficiency [1,8,10,11]. Existing methods for removing dyes from water

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have drawbacks in terms of cost, time, energy consumption, technical feasibility, and environmental concerns. Conversely, adsorption has become a preferred method for organic dye removal due to its superior efficiency, cost-effectiveness, and operational simplicity. Among the various adsorbents – such as activated carbon, zeolites, clays, resins, chitosan, and metal-organic frameworks – activated carbon is widely regarded as the benchmark, owing to its remarkably high surface area, stability, and durability [12,13]. Despite its superior adsorption capacity, the high cost of synthesis and the challenges associated with regenerating the material limit its scalability for large-scale applications. Consequently, recent research has shifted toward alternative adsorbents that are more cost-effective, efficient, and environmentally sustainable, aiming to overcome those limitations [14].

Recently, geopolymer materials have gained researchers' interest as a promising alternative for pollutant removal via adsorption. Geopolymer is an innovative green material with diverse applications, largely influenced by its structural characteristics [15,16]. It stands out as an economically attractive option due to its ability to utilize various agricultural and industrial wastes as precursors in its synthesis. Geopolymers are aluminosilicate-based materials synthesized through the activation of industrial by-products using alkaline solutions [17-19]. Their porous structure, surface functionality, and ability to be tailored for specific applications make them promising low-cost and sustainable adsorbents for environmental remediation, particularly in the removal of organic dyes from wastewater [20,21]. However, despite their promising attributes, the application of geopolymers in real-world wastewater systems remains underexplored.

Although several reviews have explored the general applications of geopolymers in environmental remediation, most of them have focused either on synthesis techniques or broad pollutant categories. Besides, existing studies often focus on laboratory-scale experiments with limited evaluation of the operational factors that influence adsorption performance. Therefore, this paper examines the synthesis and application of geopolymer materials for organic dye adsorption, with a particular focus on operational parameters. Key parameters such as solution pH, initial temperature, dye concentration, and adsorbent dosage play a vital role in optimizing their performance and improving their effectiveness in real-world wastewater treatment systems, as illustrated in Fig. 1. Expanding studies in this area is essential to fully realize the potential of geopolymers as effective adsorbents for organic dye removal. Additionally, the review highlights current research progress and identifies challenges and future directions for optimizing geopolymers as sustainable solutions in wastewater treatment. This targeted approach offers a practical framework for optimizing geopolymer performance under real-world wastewater treatment conditions. Accordingly, this review serves as a valuable resource for developing cost-effective, environmentally sustainable adsorbents. By leveraging geopolymers, this approach offers a dual advantage: promoting efficient waste management by utilizing industrial by-products and advancing sustainable wastewater treatment solutions for removing organic dyes and other pollutants.

Fig. 1. Operational Factors Affecting the Efficiency of Geopolymer Adsorbents in Wastewater Treatment

2. Classification of organic dyes

Organic dyes are essential in numerous industries, including textiles, cosmetics, plastics, and printing, and are extensively used in daily life. Their widespread application is due to their low production cost, vibrant color properties, and high resistance to environmental conditions [22]. However, these dyes tend to persist in the environment because of their stability and resistance to degradation, making them detectable across various ecosystems [23]. Furthermore, dyes are categorized into two main types – natural and synthetic, based on their sources. Natural dyes, derived from plants, insects, and animals, are limited in availability and tend to be unstable for large-scale use. Annually, about 800,000 tons of synthetic dyes are produced worldwide, and approximately 75% of this output is consumed by textile manufacturing processes [24]. However, these dyes often end up in industrial waste and are subsequently released into water bodies, contributing to environmental pollution.

The presence of organic dyes in water sources raises serious environmental and health concerns, even at low concentrations, due to their toxic nature [25]. TABLE 1 displays the worldwide standards for discharge dye effluent. Before being released into the environment, the levels of each parameter in organic dye effluent must meet the specified standards to minimize environmental impact and ensure compliance with regulatory requirements. This highlights the importance of effective treatment processes to reduce the harmful effects of organic dye pollution.

These organic dyes often have complex aromatic molecular structures, such as aromatic rings, chromophores and auxochrome that enhance their stability and resistance to microbial and chemical breakdown, making biological and physicochemical treatments less effective [26]. The discharge of these organic dyes into the environment leads to pollution and presents serious risks to aquatic organisms, microorganisms, and humans, as they have the potential to induce mutations and cancer [27]. To mitigate these risks, it's essential to remove organic dyes from wastewater before discharge. Traditional methods for dye removal include adsorption, photocatalytic degradation, reverse osmosis, coagulation, flocculation, membrane separation, and biological treatments [28]. Although some of these methods are effective, their large-scale industrial implementation is often restricted by high costs, energy consumption, sludge production, chemical usage, and limited removal efficiency. For decades, various adsorbent materials, including activated carbon, zeolite resin, chitosan, and clay minerals, have been utilized for water purification. Among these, activated carbon stands out as a highly effective adsorbent material for water remediation, owing to its exceptional adsorption capabilities. Due to the high production costs and energy-intensive regeneration of activated carbon, low-cost adsorbent materials such as geopolymers have garnered research interest as cost-effective, efficient, and environmentally sustainable alternatives for the removal of organic dyes [29].

TABLE 1

Discharge standard of dye effluent into the environment [70]

Indicator	Units	Permissible limits
Temperature	°C	40
pH	—	6.0-9.0
Biochemical oxygen demand	mg/L	250
Chemical oxygen demand	mg/L	500
Total suspended solid	mg/L	300
Total nitrogen	mg/L	50
Total phosphorus	mg/L	10
Oil and greese	mg/L	50
Ammonical nitrogen	mg/L	30

3. Geopolymerization

Geopolymers are inorganic polymers that are synthesized by the geopolymerization process, which involves the activation of alumino-silicate precursors using the alkali-activated material at room temperature or high temperature [30,31]. This process results in the formation of a three-dimensional silico-aluminate network structure, where silicon (Si) and aluminum (Al) atoms are tetrahedrally coordinated and share oxygen atoms, creating a robust structure [32,33]. This structure is derived from the tetrahedral arrangement of silicate (SiO_4) and aluminate (AlO_4) where, SiO_4 tetrahedra contribute to silicate bonds (Si-O-Si) and AlO_4 tetrahedra introduce negative charges due to the lower valence of aluminum compared to silicon, which are balanced by alkali cations like sodium (Na^+) or potassium (K^+). The classification of geopolymers depends on the Si/Al ratio in their structure. Based on this ratio, geopolymers can be categorized into three classes: polysialate, polysialate-siloxo, and polysialate-disiloxo [30]. The presence of a polymeric Si-O-Al framework is a defining feature of both geopolymers and zeolites, endowing them with shared characteristics like chemical stability and ion exchange capacity. While zeolites are crystalline and geopolymers are amorphous at room temperature, this difference gives geopolymers distinct mechanical and thermal properties, making them more suitable for structural and industrial applications. Moreover, geopolymers boast a unique combination of structural, chemical, and textural properties that enables them to effectively adsorb and immobilize heavy metals, organic pollutants, and other contaminants. Geopolymers typically feature mesopores with sizes ranging from 10 to 50 nm, which lie between micropores (<2 nm) and macropores (>50 nm). These mesopores facilitate effective diffusion and trapping of metal ions, pollutants, or other molecules within the structure [34,35].

Additionally, the formation of geopolymers follows a well-defined sequence of reactions that consists of three stages: the dissolution of aluminosilicate precursors, gel formation, and subsequent hardening [36]. Every stage is essential in influencing the final material properties of the geopolymer, including its mechanical strength, porosity, and chemical stability First, the dissolution of the aluminosilicate raw materials, such as fly ash, metakaolin, slag, or other alumina-silica-rich materials are mixed with an alkaline activator, typically sodium hydroxide (NaOH) [37]. The alkaline solution aids in the disintegration of the aluminosilicate structure, dissolving the SiO_4 and AlO_4 tetrahedra are transformed into soluble complexes [29,38,39]. Then, these complexes diffuse from the surface into the spaces between particles, forming a gel system where polymerization reactions take place. Finally, as water gradually evaporates from the gel phase, consolidation and hardening occur, leading to the formation of geopolymers with desirable mechanical properties.

The properties of geopolymers, such as surface area, pore structure, and surface charge, are largely determined by their synthesis conditions and raw materials, which directly influence the adsorption performance of the material. However, the effectiveness of these materials in removing organic dyes from wastewater also depends on various external operational parameters. The following section discusses these key parameters in detail, highlighting their roles in optimizing dye removal efficiency

4. Key factors for dye removal by geopolymer

The primary operating parameters that significantly impact the adsorption-photodegradation process include the initial concentration of organic dye, dosage of adsorbents, solution temperature, and solution pH. These factors will be thoroughly examined during the adsorption-photodegradation process, as they are essential in influencing the efficiency of organic dye removal from wastewater. In summary, a comprehensive understanding of these parameters is essential for optimizing operational conditions and maximizing the performance of geopolymer-based adsorbents in wastewater treatment applications. The following sections provide a detailed discussion of these aspects.

4.1. Initial concentration of organic dye

The adsorption capacity of organic dye is significantly affected by its initial concentration, as the removal efficiency relies on the direct interaction between organic dye molecules and the adsorption sites [40]. In general, adsorption capacity decreases as the initial dye concentration increases due to the saturation of active sites on the adsorbent surface [41]. At lower dye concentrations, more active sites are available for adsorption, whereas at higher concentrations, the limited availability of these sites constrains further adsorption. However, if the adsorption sites are not yet fully occupied, a higher initial dye concentration can improve the adsorbent's uptake capacity. This occurs because elevated dye concentrations create a stronger driving force for mass transfer, enhancing the adsorption process [42].

Açışlı, et al. [43] developed a fly ash-based geopolymer for the removal of anionic acid blue 185, with an initial dye concentration ranging from 10 to 50 mg/L, to determine its optimal performance under a 2.0 g/L geopolymer dosage at 20°C. The study found that adsorption efficiency declined as dye concentration increased. Specifically, a high removal efficiency of 87.33% was recorded at an initial concentration of 10 mg/L. This decline in efficiency is attributed to the gradual saturation of available adsorption sites as dye concentration rises. At higher concentrations, the limited number of active sites becomes occupied more quickly, restricting further adsorption and reducing overall removal efficiency. Additionally, while surface modifications have shown promising improvements in adsorption efficiency, the long-term stability, regeneration ability, and real wastewater performance of these modified materials remain underexplored.

Additionally, Shikuku et al. [44] synthesized geopolymers from volcanic ash and metakaolin for the removal of methylene blue (MB) dye. The impact of initial MB concentration on adsorption efficiency at equilibrium was examined within a range of 10–50 mg/L. Results revealed that the amount of MB adsorbed onto the solid phase at equilibrium increased significantly with higher MB concentrations. For example, the adsorption capacity of the sorbent rose from 4.8 to 24.1 mg/g as the initial MB concentration increased from 10 to 50 mg/L. Nonetheless, the study emphasizes high adsorption efficiency within a short duration but fails to evaluate the regeneration potential and long-term performance of the geopolymer adsorbents. Moreover, similar adsorption trends have been reported in other studies, aligning with our previous findings [4547].

4.2. Dosage of adsorbent

Optimizing the minimum adsorbent dosage while maximizing adsorption capacity has significant economic benefits for organic dye removal [48]. Extensive studies have been conducted to explore the relationship between adsorbent dosage and dye uptake efficiency across different systems. Generally, dye removal efficiency increases with higher adsorbent dosages, as a greater number of adsorption sites become available on the adsorbent surface. At lower dosages, adsorption occurs rapidly due to the abundance of unoccupied active sites. However, at higher dosages, the interaction between dye molecules and adsorption sites becomes kinetically restricted, requiring equilibrium to be reached before maximum adsorption can occur [49,50]. Therefore, a comprehensive investigation into the impact of adsorbent dosage provides critical insights into the feasibility of attaining optimal dye removal with minimal material input. Such an analysis is paramount in evaluating the economic viability and practical applicability of an adsorbent within large-scale treatment frameworks.

Research conducted by Onyango et al. [3] investigated the adsorption capabilities of MB dye using phosphate geopolymers derived from medical waste incinerator fly ash. The study examined the impact of varying adsorbent dosages between 0.1 g and 0.6 g on the removal of a 40 mg/L MB solution. The findings revealed that the percentage removal of MB increased concurrently with the increase in adsorbent dosage. Conversely, the adsorption capacity decreased as the adsorbent mass increased. This phenomenon is linked to the increased availability of active sites on the adsorbent as its dosage rises, while the reduction in adsorption capacity is hypothesized to result from the agglomeration of adsorption sites, restricting their accessibility. A comparable trend was identified in research on the removal of malachite green (MG) dye using laterite-rice husk ash-based geopolymers [48]. One major limitation is the lack of detailed in regeneration or reusability testing, leaving the long-term applicability and economic feasibility of the adsorbent uncertain.

Similarly, Asim et al. [32] formulated eco-friendly geopolymers from metakaolin by integrating an activator solution containing sodium silicate and sodium hydroxide to eliminate MB. Their study demonstrated that the percentage removal of MB improved when the initial adsorbent dosage was increased from 0.05 g to 0.1 g, likely due to a notable rise in active sites. However, further increasing the adsorbent dosage led to a decline in MB removal, which can be attributed to the overlapping of adsorption sites, ultimately diminishing the number of effective sites available for adsorption. Nevertheless, there was an inadequate investigation into the structural stability of the adsorbent at higher dosages, particularly regarding agglomeration effects or pore blockage that may influence adsorption kinetics. A significant limitation was the absence of reusability or desorption tests, which are crucial for assessing the practical applicability of the geopolymer in continuous or cyclic wastewater treatment systems.

Additionally, alkali-activated materials (AAMs) derived from laterite (LA) and rice husk ash (RHA) were synthesized for the adsorption of MG dye from aqueous solutions. The adsorption capacity of these materials exhibited a decreasing trend as the adsorbent dosage increased from 0.05 to 0.3 g, beyond which the values stabilized. The removal efficiency showed improvement as the adsorbent dosage increased from 0.05 to 0.1 g, a trend likely influenced by the gradual saturation of active sites and the agglomeration of adsorbent particles as the dosage increased. Excessive adsorbent mass leads to ineffective utilization of adsorption sites due to particle clustering, limiting their accessibility [51]. The substantial reduction in binding site accessibility caused by agglomeration suggests that optimal adsorption performance is achieved at adsorbent dosages below 0.3 g, with 0.1 g being the most effective for the studied alkali-activated adsorbents. Notably, while the effect of adsorbent dosage was evaluated, the study did not conduct a detailed thermodynamic assessment at varying dosages, which could provide insight into how enthalpy and entropy vary with increased adsorbent mass. Besides, the absence of a reusability and regeneration assessment leaves a critical gap in understanding the durability and lifecycle performance of the adsorbents.

4.3. Temperature

Temperature is a crucial factor in physical and chemical adsorption processes, significantly influencing the adsorbent capacity. As temperature increases, molecules move faster and viscosity decreases, resulting in higher collision frequencies between molecules [52]. Moreover, elevated temperatures lead to expanded pore volume and porosity on the adsorbent surface, ultimately enhancing adsorption-photodegradation efficiency. As a result, the temperature of the aqueous solution significantly influences the adsorption process. If the adsorption capacity increases with rising temperature, the process is considered endothermic, implying enhanced mobility of dye molecules and greater availability of active sites on the adsorbent at elevated temperatures [53,54]. On the other hand, if adsorption capacity declines with increasing temperature, the process is classified as exothermic, indicating a reduction in the strength of the interaction between dye molecules and active sites on the adsorbent as the temperature rises [55].

In 2024, [56] investigated the removal of MB dye using porous metakaolin-based geopolymer. The results indicated that adsorption capacity increased with temperature, ranging from 35°C to 45°C, suggesting an endothermic adsorption process. This observation was consistent with thermodynamic analysis, which confirmed the spontaneity of the adsorption process, as evidenced by a negative $\Delta G°$ value and a positive $\Delta H°$ value. Despite confirming the endothermic nature of the process, the study did not include kinetic modelling to determine how temperature influences adsorption rates and reaction pathways. Furthermore, the efficiency of MB removal was studied by using fly ash-based geopolymer. The key finding of this research is the significant impact of temperature on adsorption performance, where an increase in temperature within the range of 25°C to 55°C led to higher dye removal efficiency, with the maximum removal reaching approximately 96% at elevated temperatures. This effect is attributed to enhanced dye molecule mobility, faster diffusion, and increased availability of active sites at higher temperatures. However, this research lacks a comprehensive assessment of the reusability of the geopolymer adsorbent and its thermal stability during repeated adsorption-desorption cycles. These factors are crucial for real-world wastewater treatment, where adsorbents must maintain their structural integrity and effectiveness over time [57].

In a separate study, Brahmi et al. [58] developed Hydroxyapatite-metakaolin geopolymer (HAPMK-GP) pellets for the removal of brilliant green (BG) dye from aqueous solutions. As the temperature increased from 298 K to 338 K, the time required to achieve adsorption equilibrium decreased. At elevated temperatures, the rate constant (k_2) rose significantly due to an increase in the kinetic energy of molecules, which improved collision efficiency between BG dye and the granule surface, accelerating the adsorption process. Furthermore, these observations were attributed to the higher kinetic energy of BG molecules at elevated temperatures, which improved molecular collisions and reduced the thickness of the diffusion boundary layer. However, the structural stability of the HAPMK-GP under thermal influence was not evaluated. Elevated temperatures could potentially alter the material's pore structure, surface chemistry, or crystallinity, affecting long-term adsorption performance. To address this gap, comprehensive structural characterization after thermal exposure using techniques like BET, SEM, XRD, and FTIR is essential to assess the impact of elevated temperatures on adsorbent stability.

4.4. pH

Solution pH is a key factor that directly impacts adsorption efficiency by influencing the hydrogen ion concentration in the medium [52]. Changes in pH alter the electrostatic charges, ionization levels, and surface chemistry of both adsorbents and dye molecules. At low pH, the adsorbent surface becomes positively charged, enhancing electrostatic attraction with anionic dyes and increasing their adsorption efficiency [28]. However, in the case of cationic dyes, adsorption efficiency declines due to electrostatic repulsion. Conversely, at elevated pH levels, the adsorbent surface acquires a negative charge, facilitating the adsorption of cationic dyes while diminishing the efficiency for anionic dyes [59]. The point of zero charge (pHpzc) is a key factor in understanding the adsorption mechanism. It represents the pH at which the adsorbent surface has a neutral charge and is widely used to assess electrokinetic properties. Numerous studies have explored the pHpzc of various adsorbents to gain deeper insights into their adsorption behavior [55]. When the solution pH exceeds the pHpzc, the adsorbent surface becomes negatively charged due to functional groups such as OH^- and COO^-, making it more favorable for cationic dye adsorption. On the other hand, when the pH falls below the pHpzc, the adsorbent surface carries a positive charge, enhancing the adsorption of anionic dyes [60].

A study was conducted to examine the effect of pH on the removal of MB dye using foamed metakaolin-based geopolymer. The foamed structure enhances porosity and surface area, resulting in a high dye removal efficiency of 95% removal for concentrations up to 25 mg/L and a maximum uptake of 40 mg/g in batch tests. The study also investigates the effect of pH, noting that the point of zero charge (pH_pzc) of the geopolymer is approximately 8.2. Below this value, the surface tends to be positively charged, which is less favorable for adsorbing cationic dyes such as methylene blue. In contrast, pH values above 8.2 promote a negatively charged surface, enhancing electrostatic attraction and dye uptake. The findings indicate that alkaline conditions enhance the adsorption of cationic dyes like methylene blue due to favourable electrostatic interactions between the negatively charged geopolymer surface and the positively charged dye molecules [61]. Despite its promising performance, the study does not address the stability of the geopolymer under prolonged operational conditions, raising concerns about potential structural degradation or a decline in adsorption efficiency over time.

Furthermore, another study investigated this factor using a partially dealuminated metakaolin (PDK) for MB dye removal, revealing that the highest dye removal capacity of 8 mg/g at an initial dye concentration of 60 mg/L and within a pH range of 7 to 12 [62]. The results revealed that the highest dye removal occurred at alkaline pH, particularly around pH 12, where the geopolymer surface is more negatively charged and thus more effective at adsorbing the cationic MB dye. The observed improvement in removal efficiency is largely attributed to electrostatic forces, which promote the adsorption of cationic dye molecules under higher pH conditions. Nevertheless, the maximum adsorption capacity reported is relatively low compared to other geopolymer adsorbents, suggesting a need for further material optimization. Similarly, a cost-effective and efficient coal fly ash porous geopolymer (CFPG) was evaluated for Rhodamine B (RhB) removal. The effect of pH on RhB removal using CFPG was tested at pH levels of 2, 7, and 10, with a consistent removal efficiency of 100% observed across all conditions due to the zwitterionic nature of RhB [63]. However, this research does not address the desorption and regeneration of the geopolymer, which is critical for evaluating its long-term applicability and cost-effectiveness.

5. Mechanisms of dye adsorption

The adsorption of organic dyes onto geopolymer surfaces involves a range of physicochemical interactions that govern the overall removal efficiency. These adsorption mechanisms are closely linked to both the structural characteristics of the geopolymer and the external operational parameters, including adsorbent dosage, pH, temperature, and dye concentration. Understanding these underlying interactions is essential for optimizing adsorbent design and improving adsorption performance. The primary mechanisms responsible for organic dyes removal include electrostatic attraction, hydrogen bonding, ion exchange, van der Waals forces, and π–π interactions as illustrated in Fig. 2 [48]. Each mechanism plays a variable role depending on the nature of the dye, the surface chemistry of the geopolymer, and the conditions under which adsorption occurs.

Electrostatic attractions similarly have a significant impact, particularly when the dye molecules and the geopolymer surface possess opposite charges. This interaction is strongly influenced by solution pH, which determines the ionization state of dye molecules and the surface charge of the geopolymer. For instance, at low pH, the surface may become positively charged, enhancing the adsorption of anionic dyes through electrostatic attraction, whereas at high pH, cationic dyes are more effectively adsorbed [43]. In addition, chemical processes like ion exchange, complexation, and chelation additionally improve dye retention, with functional groups such as hydroxyl or silicate groups on the surface of the geopolymer establishing connections with the dye molecular structures [41]. Moreover, alterations to the surface, including the inclusion of metal oxides or organic additives, enhance the adsorption capability by boosting the surface charge density and generating additional active sites [64]. The formation of a surface complex is a key mechanism in the adsorption process, involving the binding of ions to various functional groups on the adsorbent's surface and the electrostatic interactions between the adsorbent and adsorbate surfaces [65].

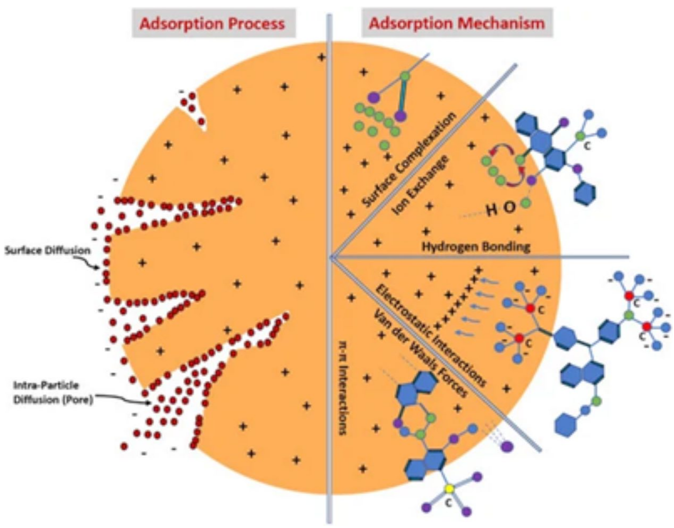

Fig. 2. Possible adsorption mechanisms for dye removal using geopolymers [48]

Other than that, hydrogen bonding occurs between the hydrogen atoms of functional groups on the geopolymer surface and electronegative atoms in the organic dye molecules. Specifically, this interaction is enhanced when the geopolymer possesses abundant surface hydroxyl groups, as pH affects the ionization of these functional groups [66]. Although van der Waals forces contribute minimally to dye adsorption compared to other mechanisms, their role becomes more significant in geopolymers with high surface area and mesoporous frameworks. These structural features enhance dye accessibility and promote greater physical interaction between the adsorbent and the dye molecules [67]. Additionally, π–π interactions occur with aromatic organic dye molecules that can align with aromatic or unsaturated regions of the geopolymer surface, such as those introduced through organic modifiers or carbonaceous residues [68]. These interactions are particularly relevant in neutral pH environments and can be enhanced by surface modifications that increase π-electron density. The temperature also affects these interactions, as higher temperatures can improve dye mobility and enhance molecular stacking, strengthening π–π interactions.

In summary, a clear grasp of electrostatic attraction, ion exchange, hydrogen bonding, van der Waals forces, and π–π interactions lays the groundwork for designing efficient geopolymer adsorbents. Therefore, building upon this mechanistic insight, numerous studies have explored the practical application of geopolymer-based materials for dye removal in wastewater treatment. Consequently, the following section reviews recent advancements, highlighting adsorption performance, key operating conditions, and the suitability of these materials for real-world implementation.

6. Adsorption of organic dyes using geopolymers

This section highlights recent research on dye adsorption using different types of geopolymer-based materials, emphasizing their adsorption capacities, influencing parameters, and modelling approaches to evaluate performance as summarized in TABLE 2. For instance, a geopolymer derived from metakaolin has been synthesized for the removal of MG dye from aqueous solutions. The findings verified the successful formation of the geopolymer, characterized by a well-developed amorphous mesoporous structure. Experimental results indicated that dye adsorption adhered to the Langmuir isotherm, achieving a maximum capacity of 185.18 mg/g. The adsorption mechanism was determined to be chemisorption, following a pseudo-second-order kinetic model. Moreover, the results demonstrated that the adsorption process is both spontaneous and endothermic, as confirmed by thermodynamic parameter analysis. The study also examined the effect of adsorbent dosage on MG dye removal, revealing an increase in removal efficiency from 47% to 97% when the geopolymer dosage was raised from 0.005 g to 0.2 g. This enhancement was attributed to the greater availability of binding sites and surface area with higher adsorbent quantities. Additionally, the geopolymer exhibited strong reusability, maintaining a substantial portion of its adsorption efficiency across five cycles. These findings highlight the potential of metakaolin-based geopolymer as an efficient, cost-effective, and environmentally sustainable adsorbent for wastewater treatment and dye removal [69]. However, the structural stability of the geopolymer over time and under repeated use was not assessed and the basic adsorption mechanism remains unexplored.

Furthermore, a novel metakaolin-based geopolymer foam adsorbent was developed using a chemical foaming method to remove MB dye from aqueous solutions. This synthesized adsorbent exhibited outstanding MB removal efficiency, achieving a maximum uptake of 90% and an adsorption capacity of 12.5 mg/g for a 10 mg/L dye solution at 40°C and pH 13. The surface morphology of the adsorbent featured a distinctive bulk porous structure with flower-like hierarchical microstructures formed by multiple nanorod-based building units, significantly enhancing MB adsorption. Thermodynamic analysis confirmed that the adsorption process was both spontaneous and endothermic, as indicated by negative $\Delta G°$ and positive $\Delta H°$ values. Additionally, the geopolymer demonstrated a strong affinity for cationic dyes due to its surface charge characteristics. These results highlight the potential of metakaolin-based geopolymer foams as an efficient, cost-effective, and environmentally sustainable solution for wastewater treatment [56]. Nonetheless, this research has limited insight into the adsorption mechanism, as it lacks kinetic and isotherm models.

Furthermore, metakaolin-based geopolymer microspheres were created by incorporating magnesium oxide (MgO) to amplify their MB adsorption capacity. The optimally formulated MK@3MgO/150-18 demonstrated a highly porous architecture with a specific surface area of 63.69 m^2/g and a maximum MB adsorption capacity of 115.70 mg/g, far surpassing microspheres without MgO. Electromagnetic interactions dominated the adsorption process, and the material showed an excellent reusability profile, retaining 95.94% of its adsorption efficacy following five consecutive cycles. Kinetic analysis indicated that the adsorption process followed a pseudo-second-order model, demonstrating that the adsorption rate is predominantly influenced by the availability of active sites on the geopolymer. Additionally, thermodynamic evaluation confirmed that the process is both endothermic and spontaneous, implying that higher temperatures improve MB adsorption, thereby enhancing the material's suitability for real-world wastewater treatment applications [28]. Although the material demonstrates good adsorption performance, the long-term structural stability and potential leaching of MgO or geopolymer components into treated water remain unexplored.

In 2024, Bassam et al. [68] designed and analysed two types of geopolymers – GPP (geopolymer powder) and Alg/GPP (geopolymer beads modified with sodium alginate) to capture methyl orange (MO) dye from aqueous solutions. The characterization results confirmed the existence of alumino-silicate gel, indicating successful geopolymerization, with notable pore volumes of 0.17 cm^3/g for GPP and 0.48 cm^3/g for Alg/GPP. The specific surface areas measured 73.56 m^2/g for GPP and 91.24 m^2/g for Alg/GPP, demonstrating effective dye adsorption. Optimal conditions for MO removal were identified as 120 mg/L for GPP in 55 minutes and 180 mg/L for Alg/GPP in 20 minutes at designated pH levels. Adsorption studies indicated that the pseudo-second-order model accurately described the retention mechanism, with chemisorption playing a dominant role. The maximum adsorption capacities, as determined by the Langmuir model, were 94.78 mg/g for GPP and 224.85 mg/g for Alg/GPP. Thermodynamic evaluations confirmed the adsorption process to be endothermic, spontaneous, and favourable, highlighting its potential for practical wastewater treatment applications. However, without assessing regeneration, it remains unclear whether the adsorbent can maintain performance over successive cycles.

TABLE 2

Comparative Analysis of Geopolymer-Based Adsorbents for Dye Removal

Study	Adsorbent	Dye	Percentage removal (%)	Adsorption capacity (mg/g)	Optimal parameter	Isotherm model	Kinetic model
Lazaryousefi et al. [69]	Metakaolin-based geopolymer	MG	97	185.18	pH 12, 40°C, 50 mg/L, 120 min	Langmuir	Pseudo-second-order
Eshghabadi et al. [56]	Metakaolin-based geopolymer foam	MB	90	12.5	pH 13, 40°C, 10 mg/L, 60 min	Not reported	Not reported
Mo et al. [28]	MK@3MgO/150-18	MB	99.62	115.70	pH 11, 30°C, 100 mg/L, 160 min	Not reported	Pseudo-second-order
Bassam et al. [68]	GPP / Alg-GPP	MO	~90-100%	94.78 (GPP), 224.85 (Alg-GPP)	GPP: pH 5, 45°C, 120 mg/L, 55 min; Alg-GPP: pH 5, 45°C, 180 mg/L, 20 min	Langmuir	Pseudo-second-order

7. Conclusion and future work

The reviewed studies highlight the significant potential of geopolymers synthesized from industrial by-products as efficient and sustainable adsorbents for eliminating organic dyes from wastewater. Besides, this review offers a comprehensive on recent advancements, with a distinct emphasis on the role of key operational parameters such as initial concentration, dosage of adsorbents, temperature, and pH in influencing adsorption efficiency. However, several research gaps remain that must be addressed to advance their practical application. Firstly, most existing studies are conducted using synthetic dye solutions under controlled laboratory conditions, which do not accurately reflect the complexity of real industrial wastewater. Future research should therefore focus on evaluating the performance of geopolymer adsorbents in real effluent systems containing multiple contaminants such as salts, surfactants, and heavy metals. According to the literature reviewed, experimental conditions such as initial concentration, dosage of adsorbents, temperature, and pH play crucial roles in the adsorption process. The current studies focus on varying individual operational parameters, such as pH, temperature, and adsorbent dosage, without considering their combined effects. Future research should employ multivariate optimization approaches, such as response surface methodology or AI-based modeling, to better understand parameter interactions.

Moreover, the adsorption mechanisms for various organic dyes include physical adsorption, electrostatic interactions, and chemisorption. Despite this known versatility, the precise contribution of each adsorption mechanism under varying operational conditions remains unclear. Most studies rely on basic isotherm and kinetic models, which do not provide detailed insight into the dominant adsorption pathways. Further research using advanced characterization techniques and molecular simulations is needed to better understand the interaction mechanisms and tailor geopolymer surfaces for improved selectivity and efficiency in dye removal. Additionally, long-term stability, regeneration, and reusability studies should be expanded to evaluate the economic feasibility of these materials on an industrial scale. Combining geopolymers with complementary treatment methods, such as photocatalysis or membrane separation, may provide synergistic effects that enhance overall treatment efficiency. Finally, comprehensive life-cycle assessments and techno-economic analyses are necessary to confirm the environmental and economic viability of geopolymer-based adsorbents compared to conventional materials. Addressing these challenges will be crucial for realizing the full potential of geopolymers in real-world wastewater treatment and promoting their implementation as green and cost-effective alternatives.

REFERENCES

[1] A. Khan, P. Ju, Z. Han, C. Ni, A Comprehensive Review on Adsorptive Removal of Azo Dyes using Functional Materials. AQUA **73** (2), 266-285 (2024).
DOI: https://doi.org/10.2166/aqua.2024.292

[2] A. Saravanan, P. Senthil Kumar, S. Jeevanantham, S. Karishma, B. Tajsabreen, P.R. Yaashikaa, B. Reshma, Effective Water/ Wastewater Treatment Methodologies for Toxic Pollutants Removal: Processes and Applications Towards Sustainable Development. Chemosphere **280**, 130595 (2021).
DOI: https://doi.org/10.1016/j.chemosphere.2021.130595

[3] C. Onyango, W. Nyairo, B. Kwach, V. Shikuku, T. Sylvain, H.D. Tamaguelon, C. Ruscher, Synthesis of Pumice and Medical Waste Incinerator Fly Based Phosphate Geopolymer for Removal of an Anionic Dye: Parameters and Adsorption Mechanism. Chemosphere **295**, 133870 (2022).
DOI: https://doi.org/10.1016/j.chemosphere.2022.133870

[4] N.N. Ahmad, A.A. Ahmad, A. Khasri, Effective Removal of Methylene Blue from Aqueous Solution by Adsorption onto Gasification Char: Isotherm, Kinetic and Thermodynamics Studies. Desalination Water Treat. **285**, 264-273 (2023). DOI:
https://doi.org/10.5004/dwt.2023.29274 10.5004/dwt.2023.29274

[5] S. Sudarshan, S. Harikrishnan, G. RathiBhuvaneswari, V. Alamelu, S. Aanand, A. Rajasekar, M. Govarthanan, Impact of Textile Dyes on Human Health and Bioremediation of Textile Industry Effluent Using Microorganisms: Current Status and Future Prospects. J. Appl. Microbiol. **134** (2), lxac064 (2023).
DOI: https://doi.org/10.1093/jambio/lxac064

[6] S.R. Satpathy, S. Bhattacharyya, Adsorptive dye removal using clay-based geopolymer: Effect of activation conditions on geopolymerization and removal efficiency. Mater. Sci. Eng. B **319**, 118348 (2025).
DOI: https://doi.org/10.1016/j.mseb.2025.118348

[7] G.A. Tochetto, L. Simão, D. de Oliveira, D. Hotza, A.P.S. Immich, Porous Geopolymers as Dye Adsorbents: Review and Perspectives. J. Clean. Prod. **374**, 133982 (2022).
DOI: https://doi.org/10.1016/j.jclepro.2022.133982

[8] M. Novotná, P. Knotek, T. Hanzlíček, P. Kutálek, I. Perná, K. Melánová, E. Černoškováa, K. Kopecká, TiO_2 Modified Geopolymers for the Photocatalytic Dye Decomposition. Crystals **11** (12), 1511 (2021). DOI: https://doi.org/10.3390/cryst11121511

[9] L. Kemdjien, A. Tchakounte, I. Lenou, S. Tome, C. Kede, Optimization of synthesis of a geopolymer based on laterite/beer bottles composites for adsorption of methylene blue in aqueous solution. Desalin. Water Treat. **322**, 101189 (2025).
DOI: https://doi.org/10.1016/j.dwt.2025.101189

[10] D. Glažar, B. Simončič, TiO_2 and ZnO as Advanced Photocatalysts for Effective Dye Degradation in Textile Wastewater. Tekstilec. **66** (3), 178-198 (2023).
DOI: https://doi.org/ 10.14502/tekstilec.66.2023045

[11] R. Agarwala, L. Mulky, Adsorption of Dyes from Wastewater: A Comprehensive Review. Chem. Bio. Eng. Revi. **10** (3), 326-335 (2023). DOI: https://doi.org/10.1002/cben.202200011

[12] N.A. Yaacob, A. Khasri, N.H. Mohd Salleh, M.R. Mohd Jamir, Optimization of AC/TiO_2-Cu Ternary Composite Preparation with Enhanced UV Light Activity for Adsorption–Photodegradation of Metronidazole via RSM-CCD. J. Disper. Sci. Technol. **45** (2), 249-260 (2024).
DOI: https://doi.org/10.1080/01932691.2022.2143367

[13] G. Song, W. Fan, J. Zhang, Z. Liu, Y. Shi, Y. Sun, G. Ding, Insights into the adsorption behavior and mechanism of ionic dyes on carbon nanotubes with different functional groups. Diam. Relat. Mater. **157** (2025) 112577.
DOI: https://doi.org/10.1016/j.diamond.2025.112577

[14] H. Lin, J. Zhang, R. Wang, W. Zhang, J. Ye, Adsorption properties and mechanisms of geopolymers and their composites in different water environments: A comprehensive review. J. Water Process Eng. **62** (2024) 105393.
DOI: https://doi.org/10.1016/j.jwpe.2024.105393

[15] G. Furtos, D. Prodan, C. Sarosi, M. Moldovan, M. Lach, M. Melnychuk, K. Korniejenko, Advanced Geopolymer-Based Composites for Antimicrobial Application. Mater **16** (23), 1-14 (2023). DOI: https://doi.org/10.3390/ma16237414

[16] I.H. Aziz, N.A. Mazlan, M.M.A. B. Abdullah, N.F. Shahedan, R. Ediati, W.M. W Ibrahim, H. Fansuri, Kaolin-Based Geopolymer as a Heavy Metal Removal: Short Review. Eur. J. Mater. Sci. Eng. **8 (**1), 58-66 (2023).
DOI: https://doi.org/10.36868/ejmse.2023.08.01.058

[17] P. Devarajan, D.S. Vijayan, S.R. Kumar, A.F.J. King, A Short Review on Substantial role of Geopolymer in The Sustainable Construction Industry. IOP Conf. Ser. Earth Environ. Sci. **1130** (1), 7414 (2023).
DOI: https://doi.org/10.1088/1755-1315/1130/1/012002

[18] B. Primasari, W. Wazien, A. Zailani, Comparative Study of Concrete Wastes as Low-Cost Adsorbent for Copper Removal from Aqueous Solution Comparative Study of Concrete Wastes as Low-Cost Adsorbent for Copper Removal from Aqueous Solution. J. Kejuruteraan. **36**, 36-46 (2024).
DOI: https://doi.org/10.17576/jkukm-2024-36(5)-31

[19] Z. Rahmawati, P. Sánchez-Camacho, G.A. Mutch, A.A. Widati, T. Gunawan, H. Fansuri, Geopolymer membrane – photocatalyst coupling for textile wastewater treatment. Appl. Clay Sci. **275**, 107886 (2025).
DOI: https://doi.org/10.1016/j.clay.2025.107886

[20] N.F. Shahedan, T, Hadibarata, M.M.A.B. Abdullah, M.N.H. Jusoh, S.Z. Abd Rahim, I. Isia, A. Armada Bras, A. Bouaissi, F.H. Juwono, Potential of Fly Ash Geopolymer Concrete as Repairing and Retrofitting Solutions for Marine Infrastructure: A Review. CSCM. 20, e03214 (2024).
DOI: https://doi.org/10.1016/j.cscm.2024.e03214

[21] M.S. Akhtar, S. Ali, W. Zaman, Innovative Adsorbents for Pollutant Removal: Exploring the Latest Research and Applications. Molecules. **29** (18), 1-37 (2024).
DOI: https://doi.org/10.3390/molecules29184317

[22] N.A. Yaacob, A. Khasri, N.H.M. Salleh. Ridzuan, Statistical Optimization of AC/TiO_2-Cu Composite via Response Surface Methodology (RSM). J. Mech. Eng. 1**1**, (1), 199-209 (2022).
DOI: https://doi.org/10.24191/jmeche.v11i1.23598

[23] N.A. Yaacob, A. Khasri, M.R.M. Jamir, N.H.M. Salleh, M.A. Ahmad, Optimization of activated carbon modified with titanium dioxide composite preparation for adsorption/photodegradation of malachite green dye removal. AIP Conf. Proc. **2883**, 1-15 (2024).
DOI: https://doi.org/10.1063/5.0207067

[24] B. Pizzicato, S. Pacifico, D. Cayuela, G. Mijas, M. Riba-Moliner, Advancements in Sustainable Natural Dyes for Textile Applications: A Review. Molecules **28**, 1-22 (2023).
DOI: https://doi.org/10.3390/molecules28165954

[25] R. Al-Tohamy, S.S. Ali, F. Li, K.M. Okasha, Y.A.G. Mahmoud, T. Elsamahy, H. Jiao, Y. Fu, J. Sun, A critical review on the treatment of dye-containing wastewater: Ecotoxicological and health concerns of textile dyes and possible remediation approaches for environmental safety. Ecotoxicol. Environ. Saf. **231**, 113160 (2022).
DOI: https://doi.org/10.1016/j.ecoenv.2021.113160

[26] N. Muhd Julkapli, S. Bagheri, S. Bee Abd Hamid, Recent advances in heterogeneous photocatalytic decolorization of synthetic dyes. Sci. World J. **2014**, 692307 (2014).
DOI: https://doi.org/10.1155/2014/692307

[27] H. Ben Slama, A.C. Bouket, Z. Pourhassan, F.N. Alenezi, A. Silini, H. Cherif-Silini, T. Oszako, L. Luptakova, P. Golińska, L. Belbahri, Diversity of synthetic dyes from textile industries, discharge impacts and treatment methods. Appl. Sci. **11**, 1-21 (2021).
DOI: https://doi.org/10.3390/app11146255

[28] H. Mo, L. Liu, Z. Yang, Y. Lao, Y. He, The Effect of Magnesium Oxide on The Structure of Metakaolin-Based Geopolymer Microspheres and Its Application in The Adsorption of Methylene Blue Dye. J. Water Process Eng. **70**, 106937 (2025).
DOI: https://doi.org/10.1016/j.jwpe.2025.106937

[29] J. Matsimbe, M. Dinka, D. Olukanni, I. Musonda, Geopolymer: A Systematic Review of Methodologies. Mater. **15** (19), 6852 (2022). DOI: https://doi.org/10.3390/ma15196852

[30] C. Bai, K. Zheng, F. Sun, X. Wang, L. Zhang, T. Zheng, P. Colombo, B. Wang, A review on metakaolin-based porous geopolymers. Appl. Clay Sci. **258**, 107490 (2024). DOI: https://doi.org/10.1016/j.clay.2024.107490

[31] Y. Yu, F. Xu, S. Wang, L. Fan, J. Zhang, P. Li, L. Yu, Influence mechanism of curing temperature on geopolymerization reaction: A comprehensive review. J. Build. Eng. **103**, 112195 (2025). DOI: https://doi.org/10.1016/j.jobe.2025.112195

[32] N. Asim, M. Badiei, M. Shakeri, Z. Emdadi, N.A. Samsuddin, S. Soltani, N. Amin, Development of Green Photocatalytic Geopolymers for Dye Removal. Mater Chem Phys. **283**, 126020 (2022). DOI: https://doi.org/10.1016/j.matchemphys.2022.126020

[33] A.A. Siyal, R.M.S. Radin Mohamed, R. Shamsuddin, M.B. Ridzuan, A comprehensive review of synthesis kinetics and formation mechanism of geopolymers. RSC Adv. **14**, 446-462 (2024). DOI: https://doi.org/10.1039/d3ra06205h

[34] E.Y. Ngu, J.A. Tchio, L.L. Duna, C.R. Kaze, T.K. Herve, C. Leonelli, Design and Characterization of Porous Geopolymer for Dye Removal. Int. J. Waste Resour. **13**, 522 (2023). DOI: https://doi.org/10.35248/2252-5211.23.13.5

[35] S. Qaidi, H.M. Najm, S.M. Abed, H.U. Ahmed, H. Al Dughaishi, J. Al Lawati, M.M. Sabri, F. Alkhatib, A. Milad, Fly Ash-Based Geopolymer Composites: A Review of the Compressive Strength and Microstructure Analysis. Materials (Basel). **15**, 7098 (2022). DOI: https://doi.org/10.3390/ma15207098

[36] J. Xu, M. Li, D. Zhao, G. Zhong, Y. Sun, X. Hu, J. Sun, X. Li, W. Zhu, M. Li, Z. Zhang, Y. Zhang, L. Zhao, C. Zheng, X. Sun, Research and Application Progress of Geopolymers in Adsorption: A Review. Nanomaterials **12**, 3002 (2022). DOI: https://doi.org/10.3390/nano12173002

[37] Y. Ettahiri, B. Akhsassi, M. El Fazdoune, A. Bouddouch, L. Bouna, A. Benlhachemi, L. Pérez-Villarejo, R.D.F. P.M. Moreira, From Synthesis to Applications: A Comprehensive Review of Geopolymer Materials for Photocatalytic Degradation of Organic Pollutants. Sep. Purif. Technol. **330**, 125396 (2024). DOI: https://doi.org/10.1016/j.seppur.2023.125396

[38] P. Rożek, M. Król, W. Mozgawa. Lightweight Geopolymer-Expanded Glass Composites for Removal of Methylene Blue from Aqueous Solutions. Ceram. Int. **46** (12), 19785-19791 (2020). DOI: https://doi.org/10.1016/j.ceramint.2020.05.011

[39] F.F. Kamarzaman, M.M.A.B. Abdullah, M.R. Rozainy, R. Ahmad, W.M.W. Ibrahim, M.F. Omar, C.M.R. Ghazali, W.M.A.W. Ibrahim, A Comparative review on Low-Cost Adsorbent based Alkali-Activated materials by Adsorption study. Arch. Metall. Mater. **69**, 1385-1394 (2024). DOI: https://doi.org/10.24425/amm.2024.151404.

[40] Lalliansanga, D. Tiwari, A. Tiwari, A. Shukla, M.J. Shim, S.M. Lee, Facile synthesis and characterization of Ag(NP)/TiO_2 nanocomposite: Photocatalytic efficiency of catalyst for oxidative removal of Alizarin Yellow. Catal. Today **388-389**, 125-133 (2022). DOI: https://doi.org/10.1016/j.cattod.2020.09.010.

[41] M.I. Khan, S. Sufian, F. Hassan, R. Shamsuddin, M. Farooq, Phosphoric acid based geopolymer foam-activated carbon composite for methylene blue adsorption: isotherm, kinetics, thermodynamics, and machine learning studies. RSC Adv. **15**, 1989-2010 (2025). DOI: https://doi.org/10.1039/d4ra05782a

[42] Y. Ettahiri, B. Bouargane, K. Fritah, B. Akhsassi, L. Pérez-Villarejo, A. Aziz, L. Bouna, A. Benlhachemi, R.M. Novais, A state-of-the-art review of recent advances in porous geopolymer: Applications in adsorption of inorganic and organic contaminants in water. Constr. Build. Mater. **395**, 132269 (2023). DOI: https://doi.org/10.1016/j.conbuildmat.2023.132269

[43] Ö. Açışlı, İ. Acar, A. Khataee, Preparation of a Surface Modified Fly Ash-Based Geopolymer for Removal of an Anionic Dye: Parameters and Adsorption Mechanism. Chemosphere. **295**, 133870 (2022). DOI: https://doi.org/10.1016/j.chemosphere.2022.133870

[44] V.O. Shikuku, S. Tome, D.T. Hermann, G.A. Tompsett, M.T. Timko, Rapid Adsorption of Cationic Methylene Blue Dye onto Volcanic Ash-metakaolin based Geopolymers. Silicon. **14** (15), 9349-9359 (2022). DOI: https://doi.org/10.1007/s12633-021-01637-9

[45] E.M. Abdel Hamid, H.M. Aly, K.A.M. El Naggar, Synthesis of nanogeopolymer adsorbent and its application and reusability in the removal of methylene blue from wastewater using response surface methodology (RSM). Sci. Rep. **14**, 1-25 (2024). DOI: https://doi.org/10.1038/s41598-024-70284-y

[46] A. Purbasari, D. Ariyanti, E. Fitriani, Adsorption of Methyl Orange Dye by Modified Fly Ash-Based Geopolymer – Characterization, Performance, Kinetics and Isotherm Studies. J. Ecol. Eng. **24**, 90-98 (2023). DOI: https://doi.org/10.12911/22998993/157541

[47] T.P. Le, H.V.T. Luong, H.N. Nguyen, T.K.T. Pham, T.L. Trinh Le, T.B.Q. Tran, T.N.M. Ngo, Insight into adsorption-desorption of methylene blue in water using zeolite NaY: Kinetic, isotherm and thermodynamic approaches. Results Surf. Interfaces. **16**, 100281 (2024). DOI: https://doi.org/10.1016/j.rsurfi.2024.100281

[48] Y. Fang, L. Yang, F. Rao, K. Zhang, Z. Qin, Z. Song, Z.Z. Na, Behaviors and Mechanisms of Adsorption of MB and Cr (VI) by Geopolymer Microspheres under Single and Binary Systems. Mol. **29** (7), 1560 (2024). DOI: https://doi.org/10.3390/molecules29071560

[49] A. Maleki, M. Mohammad, Z. Emdadi, N. Asim, M. Azizi, J. Safaei, Adsorbent materials based on a geopolymer paste for dye removal from aqueous solutions. Arab. J. Chem. **13**, 3017-3025 (2020). DOI: https://doi.org/10.1016/j.arabjc.2018.08.011

[50] S. Yan, M. Zhang, D. Fu, C. He, S. Wang, X. Zhai, Synthesis of red mud/metakaolin based geopolymer for adsorption of methylene blue and tetracycline hydrochloride from water. Colloids Surfaces A Physicochem. Eng. Asp. **722**, 137155 (2025). DOI: https://doi.org/10.1016/j.colsurfa.2025.137155.

[51] S. Tome, V. Shikuku, H.D. Tamaguelon, S. Akiri, M.A. Etoh, C. Ruscher, J. Etame, Efficient Sequestration of Malachite Green in Aqueous Solution by Laterite-Rice Husk Ash-Based Alkali-Activated Materials: Parameters and Mechanism. ESPR. **30** (25), 67263-67277 (2023). DOI: https://doi.org/10.1007/s11356-023-27138-3

[52] H.X. Su, C.J. Li, Y.J. Zhang, Facile Synthesis of Geopolymer-Based Hierarchical Porous Materials for Efficient Adsorption-Photocatalysis of Dye Wastewater. J. Environ. Chem. Eng. **12** (6), 114809 (2024).
DOI: https://doi.org/10.1016/j.jece.2024.114809

[53] C. Sarkar, J.K. Basu, A.N. Samanta, Synthesis of Novel ZnO/Geopolymer Nanocomposite Photocatalyst for Degradation of Congo Red Dye Under Visible Light. Environ. Nanotechnol. Monit. Manag. **16**, 100521 (2021).
DOI: https://doi.org/10.1016/j.enmm.2021.100521

[54] X. Jaramillo-Fierro, S. Gaona, J. Ramón, E. Valarezo, Porous Geopolymer/$ZnTiO_3$/TiO_2 Composite for Adsorption and Photocatalytic Degradation of Methylene Blue Dye. Polymers (Basel). **15** (12), 2697 (2023).
DOI: https://doi.org/10.3390/polym15122697

[55] A.D'Angelo, V. Viola, M. Fiorentino, G. Dal Poggetto, I. Blanco, Use of Natural Dyes to Color Metakaolin-Based Geopolymer Materials. Ceram Int. **51** (5), 5528-5535 (2024).
DOI: https://doi.org/10.1016/j.ceramint.2024.05.109

[56] F. Eshghabadi, V. Javanbakht, Preparation of Porous Metakaolin-Based Geopolymer Foam as An Efficient Adsorbent for Dye Removal from Aqueous Solution. J. Mol. Struct. **1295**, 136639 (2024).
DOI: https://doi.org/10.1016/j.molstruc.2023.136639

[57] B. Aouan, S. Alehyen, M. Fadil, M. El Alouani, H. Saufi, E.H. El Herradi, F. El Makhoukhi, M. Taibi, Development and optimization of geopolymer adsorbent for water treatment: Application of mixture design approach. J. Environ. Manage. **338**, 117853 (2023).
DOI: https://doi.org/10.1016/j.jenvman.2023.117853

[58] A. Brahmi, S. Ziani, S. AitAli, B.N. Benkhaoula, Y. Yu, H. Ahouari, H. Khireddine, T. Luukkonen, Porous metakaolin geopolymer as a reactive binder for hydroxyapatite adsorbent granules in dye removal. Hybrid Adv. **5**, 100134 (2024).
DOI: https://doi.org/10.1016/j.hybadv.2023.100134

[59] A. Brahmi, S. Ziani, S. Aitali, K. Ben, H. Ahouari, T. Luukkonen, Synergistic adsorption and photocatalytic removal of hexavalent chromium and direct red 23 dye using porous magnetic hydroxyapatite-metakaolin geopolymer granules doped with TiO_2. J. Environ. Chem. Eng. **13**, 117497 (2025).
DOI: https://doi.org/10.1016/j.jece.2025.117497

[60] H. Lee, S. Fiore, F. Berruti, Adsorption of Methyl Orange and Methylene Blue on Activated Biocarbon Derived from Birchwood Pellets. Biomass Bioenergy **191**, 107446 (2024).
DOI: https://doi.org/10.1016/j.biombioe.2024.107446

[61] S. Candamano, G. Coppola, A. Mazza, J.I. Caicho Caranqui, S. Bhattacharyya, S. Chakraborty, F. Alexis, C. Algieri, Batch and Fixed Bed Adsorption of Methylene Blue onto Foamed Metakaolin-Based Geopolymer: A Preliminary Investigation. Chem. Eng. Res. Des. **197**, 761-773 (2023).
DOI: https://doi.org/10.1016/j.cherd.2023.08.014

[62] K. Elewa, A.F. Tawfic, M. Tarek, N.A. Al-Sagheer, N.M. Nagy, Removal of methylene blue from synthetic industrial wastewater by using geopolymer prepared from partially dealuminated metakaolin. Sci. Rep. **15**, 1-17 (2025).
DOI: https://doi.org/10.1038/s41598-025-01461-w

[63] E.C. Umejuru, R. Street, J.N. Edokpayi, The Application of Synthesized Geopolymer for The Removal of Cationic Dye from Industrial Wastewater. Results Mater. **22**, 100572 (2024).
DOI: https://doi.org/10.1016/j.rinma.2024.100572

[64] H. Lin, J. Zhang, R. Wang, W. Zhang, J. Ye, Adsorption Properties and Mechanisms of Geopolymers and Their Composites in Different Water Environments: A Comprehensive Review. J. Water Process Eng. **62** (2024).
DOI: https://doi.org/10.1016/j.jwpe.2024.105393

[65] A. Ben Amor, M. Arenas, J. Martín, A. Ouakouak, J.L. Santos, I. Aparicio, E. Alonso, N. Hamdi, Clays-based geopolymers: a sustainable application as adsorbent of cytostatic drugs for water purification. Appl. Water Sci. **14**, 1-14 (2024).
DOI: https://doi.org/10.1007/s13201-024-02273-5

[66] L. Zhang, X. Ling, Y. Liu, Z. Chen, B. He, Y. Wang, Z. Tang, D. Cang, Carbon negative technology empowered adsorption behaviour and mechanism of porous geopolymer. Environ. Res. **277**, 121545 (2025).
DOI: https://doi.org/10.1016/j.envres.2025.121545

[67] C.J. Li, Y.J. Zhang, H. Chen, P.Y. He, Q. Meng, Development of porous and reusable geopolymer adsorbents for dye wastewater treatment. J. Clean. Prod. 348, 131278 (2022).
DOI: https://doi.org/10.1016/j.jclepro.2022.131278

[68] A. Bassam, R. Bassam, M. El Alouani, Y. Rachdi, H. Saufi, S. Belaaouad, Synthesis of Geopolymer Powder and Beads Based on Red Clay Waste for The Adsorption of Methyl Orange Dye from Aqueous Solutions: Characterization, Application of Response Surface Methodology, and Cost Analysis. Sustain. Chem. Pharm. **39**, 101575 (2024).
DOI: https://doi.org/10.1016/j.scp.2024.101575

[69] A. Lazaryousefi, P. Najibikhah, A. Rahbar-Kelishami, Application of Meta-Kaolin Based Geopolymer for The Removal of Malachite Green Dye from Aqueous Solution: Kinetics, Isotherms and Thermodynamics Studies. J. Mol. Liq. **419**, 126800 (2025).
DOI: https://doi.org/10.1016/j.molliq.2024.126800

[70] Malaysia. Attorney General's Chambers. Water Services Industry (Prohibited Effluent) Regulations 2021 [P.U. (A) 426], Government Gazette, 25 November 2021. Available at: https://lom.agc.gov.my/ilims/upload/portal/akta/outputp/1715485/PUA426_2021.pdf