

APPLICATION OF SMART HYDROGELS IN SUSTAINABLE TREATMENT OF EFFLUENTS FROM THE TEXTILE INDUSTRY: A REVIEW

Aplicación de hidrogeles inteligentes en el tratamiento sostenible de efluentes de la industria textil. Una revisión

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ABSTRACT

Dyes are among the most harmful contaminants produced by industries such as textile, rubber, paper, and others. These contaminants are eliminated from the production process and disposed of as effluents, most of which are ultimately discharged into bodies of water such as lakes, rivers, beaches, and streams. The majority of these industrial dyes are categorized as toxic and/or carcinogenic, with their stability and ability to integrate into the ecological chain potentially affecting flora and fauna. Additionally, sustainable development goals have been proposed globally since 2015, including the implementation of eco-friendly technologies for the treatment of water resources to enable reuse. Therefore, pretreatment of these textile industrial effluents before their discharge into water bodies is imperative. This paper presents a literature review on effluent treatment using hydrogels, focusing on the textile industry. Hydrogels can serve as an adsorption technology with significant comparative advantages over other removal methods, including low cost, reusability, biodegradability, non-toxicity, absence of secondary pollutants, and ease of operational management. The compiled results underscore the capability of hydrogels in dye removal. Furthermore, their removal efficiency, kinetics, reusability cycles, thermodynamics, equilibrium, and responses to various environmental stimuli reinforce the feasibility of implementing hydrogels as an adsorption technology for treating textile industrial effluents.

Palabras clave: hidrogeles, adsorción, tratamiento de aguas, eliminación de colorantes.

RESUMEN

Los colorantes se encuentran entre los contaminantes más perjudiciales producidos por industrias como la textil, la del caucho, la papelería y otras. Estos contaminantes son eliminados del proceso de producción y dispuestos como efluentes, la mayoría de los cuales son finalmente descargados en cuerpos de agua como lagos, ríos, playas y arroyos. La mayoría de estos colorantes industriales están clasificados como tóxicos y/o cancerígenos, y su estabilidad y capacidad de integrarse en la cadena ecológica

pueden afectar la flora y la fauna. Además, desde 2015 se han propuesto, a nivel mundial, objetivos de desarrollo sostenible que incluyen la implementación de tecnologías ecológicas para el tratamiento de los recursos hídricos que permitan su reutilización. Por lo tanto, es imperativo realizar un pretratamiento de estos efluentes industriales textiles antes de su descarga en cuerpos de agua. Este artículo presenta una revisión bibliográfica sobre el tratamiento de efluentes mediante hidrogeles, con enfoque en la industria textil. Los hidrogeles pueden actuar como una tecnología de adsorción con ventajas comparativas significativas frente a otros métodos de eliminación, incluyendo bajo costo, reutilización, biodegradabilidad, no toxicidad, ausencia de contaminantes secundarios y facilidad de manejo operativo. Los resultados recopilados destacan la capacidad de los hidrogeles para eliminar colorantes. Además, su eficiencia de eliminación, cinética, ciclos de reutilización, termodinámica, equilibrio y respuesta a diversos estímulos ambientales refuerzan la viabilidad de implementar hidrogeles como tecnología de adsorción para el tratamiento de efluentes industriales textiles.

INTRODUCTION

Approximately, there are more than 100 000 industrial dyes (textile, cosmetic, food, plastic, rubber, leather, printing, paper, pharmaceutical), which have significantly increased due to scientific and technological advancements in synthesizing new dyes with specific characteristics tailored to the needs of various industries (Lellis et al. 2019, Kivanç et al. 2020). Moreover, these dyes are released during production and discarded as effluents, ultimately entering bodies of water, including lakes, rivers, beaches, streams, and more (Choudhury 2017, Flores-Valenzuela et al. 2023). Most industrial dyes are classified as toxic and/or carcinogenic. Their stability and inability to integrate into the ecological chain potentially impact the flora and fauna (Mehra et al. 2021, Soni 2021, Li et al. 2023). This stability and resistance to degradation, light, heat, and oxidation stem from their stable structure and complex aromatic chains (Eskhan et al. 2018).

In particular, the textile industry requires large amounts of water and dyes for textile production (Mondal et al. 2017), which include acid, basic, reactive, azoic, diazoic, and anthraquinone dyes (Singh and Singh 2017, Nakhjiri et al. 2018, Benkhaya et al. 2020, Tara et al. 2020). As a result, the textile industry is one of the main contributors to the production of industrial wastewater, with its dyes exhibiting a chroma composition ranging from 500 to 5 000 000 (Yang et al. 2018). Accordingly, industrial dye pollution is alarming, and its impact is significant even at low concentrations. Therefore, it is imperative to pretreat these toxic components before their discharge into water bodies (Rech and Paulino 2019, Selvaraj et al. 2021, Flores-Valenzuela et al. 2023).

From the ecological point of view, the toxicity of industrial dyes represents a significant environmental issue due to its detrimental impact on aquatic and terrestrial ecosystems. These dyes contain toxic chemical compounds (aromatic), heavy metals, and chlorides that could seep into the soil or water bodies, harming biodiversity and the health of biological communities. The stability and low biodegradability of these toxic dyes could lead to a decline in water quality (reduction in water transparency and dissolved oxygen), altering biochemical cycles (affecting photosynthesis by obstructing light penetration) and activity due to an accumulation of toxins in organism tissues, which could threaten the equilibrium of ecosystems (Rech and Paulino 2019, Selvaraj et al. 2021).

The most important class of dyes is the azo type because of its superior dyeing capacity, ease of preparation, ready availability, and ability to cover the entire chromatic range. However, their high stability and toxicity pose serious risks to animal and human health through the food chain process. These contaminants, seeping into water bodies, not only endanger aquatic life but also bioaccumulate within organisms, ultimately finding their way into the human food chain (Selvaraj et al. 2021).

One classification of industrial dyes is based on the charge of the material: (1) cationic dyes, (2) non-ionic dyes, and (3) anionic dyes (Affat 2021). Within this classification, cationic dyes are the most hazardous contaminants, surpassing anionic ones. This is because cationic dyes can readily interact with the negative charges of organic matter, forming highly stable bonds. These dyes are estimated to be produced at rates of 700 000 t/year, of which 12% are discharged untreated into the environment (Rizzi et al. 2018, Foroutan et al. 2019a).

Over the years, traditional methods for treating wastewater from the textile industry have become unreliable, obsolete, or inefficient due to the new characteristics and properties of dye chemical compounds. Hence, there is an urgent need to develop new technologies and materials capable of removing these contaminants from wastewater, not only at the laboratory level but also at an industrial scale (Priya et al. 2019, Flores-Valenzuela et al. 2023). **Table I** summarizes the main technologies currently employed for this purpose.

These new materials must fulfill technical requirements of efficiency, operability, and non-contamination. Additionally, economic factors such as accessibility and low cost are essential.

Globally, various methods for dye removal have been employed, including physical, chemical, biological, adsorption, membrane filtration, ozonation, microwave-assisted catalysis, biodegradation, photocatalytic degradation, coagulation-flocculation, chemical oxidation, electrochemical degradation, and electro-flocculation, among others (Piaskowski et al. 2018, Chen et al. 2020, Kıvanç and Yönten 2020, Wang et al. 2020, Bustos-Terrones et al. 2021, Flores-Valenzuela et al. 2023). However, most of these methods pose significant economic and technical challenges for industrial-scale implementation (Ertugay and Acar 2017, Bento et al. 2020, Li et al.

2020, Ozturk and Yilmaz 2020, Wang and Chen 2020). On the other hand, adsorption-based separation technology offers clear advantages over other methods, such as low investment, profitability, high efficiency in removing both cationic and anionic species, ease of operation, renewability, among others (García et al. 2018, Munagapati et al. 2018, Foroutan et al. 2019b, Flores-Valenzuela et al. 2023, Wang et al. 2023).

Currently, various materials have been used as adsorbents, including activated carbon, clay, biomass, carbon nanotubes, ion exchange resins, and polymers, among others (Noorimotlagh et al. 2019, Rehman et al. 2019, Staudt et al. 2020, Yönten et al. 2020, Flores-Valenzuela et al. 2023). Among the broad spectrum of adsorbents, polymeric materials offer advantages and features that make them appealing for large-scale applications (Aichour et al. 2018, Sharma et al. 2018a, Tan and Sen 2019). These characteristics include easy material availability, affordability, ease of operation, regenerability, and more.

Within the realm of polymeric materials, hydrogels stand out as prominent candidates for adsorbing contaminants in industrial water bodies, particularly in the textile industry. Among their main characteristics, two stand out as significant advantages: biodegradability and the absence of secondary pollutant agents. Biodegradability is achieved through natural polymers or modified synthetic polymers

TABLE I. KEY TECHNOLOGIES FOR INDUSTRIAL DYE REMOVAL.

Reference	Technology	Principle	Disadvantages
Kanakaraju et al. (2018)	Catalytic ozonation	Advanced oxidation	Short ozone lifespan, high energy requirement, sample contamination
Piepiórka-Stepuk (2018)	Membrane filtration	Physical separation	Short membrane lifespan, fouling, and non-regeneration
Zuo et al. (2020)	Microwave catalysis	Energetic decomposition	High investment costs and lack of secondary studies
Matafonova and Batoev (2018)	Photocatalytic degradation	UV light and TiO ₂ /ZnO catalysts	Not applicable at industrial scale
Zhao et al. (2021)	Coagulation-flocculation	Clustering separation	Use of chemicals and sludge generation
Crini and Lichtfouse (2018)	Chemical oxidation	Chemical bond breakage	High energy demand
Martínez-Huitle and Panizza (2018)	Electrochemical degradation	Electrochemical reaction	Low efficiency and high operational costs
Rech and Paulino (2019)	Electro-flocculation	Clustering via electrical current	Not applicable at industrial scale and high energy demand

featuring a hydrogel structure susceptible to degradation under specific environmental conditions and thus does not generate any contamination to the environment. The second key characteristic is the absence of secondary pollutant agents, meaning that during the hydrogel's use and once it reaches the end of its lifetime, these polymers do not generate any pollution that would harm the environment or water bodies. Other important attributes of hydrogels include non-toxicity, cost-effectiveness, ease of preparation, eco-friendliness, and recyclability (Pereira et al. 2017, van Tran et al. 2018, Chen and Zhu 2019, Samaddar et al. 2019, Verma et al. 2020).

Hydrogels are ideal adsorbents for ionic elements and compounds because of their three-dimensional structure. The polymeric chains and functional groups within hydrogels exhibit hydrophilic behavior, enabling them to adhere to dye ions for removal. Hydrogels are strong candidates for removing contaminants from the textile industry, often achieving removal efficiencies above 95% (Pereira et al. 2017, van Tran et al. 2018, Chen and Zhu 2019, Verma et al. 2020).

Furthermore, hydrogels have exhibited selectivity in the removal of both cationic dyes (Sánchez et al. 2019) and anionic dyes (Safavi-Mirmahalleh et al. 2020), either simultaneously or individually, under various operating conditions such as pH, temperature, initial dye concentration, contact time, and more (Vilela et al. 2019).

Selective adsorption enables the recovery and reuse of synthetic dyes, preventing environmental contamination. Hydrogels, driven by electrostatic forces, offer multiple reuses with sustained efficiency, rendering them sustainable for industrial effluent treatment. Their renewable capacity addresses modern pollution challenges, offering a significant advantage over alternative technologies. Hydrogels can be utilized repeatedly until their adsorption capacity is exhausted, representing a cost-effective and eco-friendly solution for textile wastewater treatment. However, pretreatment with HCl or NaOH is necessary for the regeneration of active sites (Priya et al. 2019).

KEY PARAMETERS INFLUENCING HYDROGEL ADSORPTION

Dye removal

The amount of removed dye (adsorbate), measured in milligrams of dye per gram of dry hydrogel, is calculated according to Kivanç et al. (2020) and Ferfera-Harrar et al. (2021):

$$q_e = \frac{(C_0 - C_e) \times V}{W} \quad (1)$$

where C_0 and C_e represent the initial (mg/L) and equilibrium concentrations of the dye in the solution, respectively; V indicates the volume (L) of the solution containing the dye to be removed, W is the mass (g) of the hydrogel (adsorbent), and q_e represents the amount of dye adsorbed at equilibrium (mg/g).

A simpler way to represent the amount of adsorbed dye is through the removal percentage, given by Nakhjiri et al. (2018) and Ferfera-Harrar et al. (2021):

$$\% \text{ removal} = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (2)$$

Adsorption isotherms

Isotherms model the equilibrium behavior of the adsorption process. The Langmuir and Freundlich isotherms are the most commonly used to represent dye adsorption onto hydrogels. The Langmuir isotherm is described by Dudu et al. (2017), Eskhan et al. (2018), and Kivanç et al. (2020):

$$\frac{C_e}{q_e} = \left(\frac{C_e}{q_{\max}} \right) + \left(\frac{1}{q_{\max} \times K_L} \right) \quad (3)$$

where $q_{\max}$ is the maximum amount of dye adsorbed per adsorbent (mg/g) and K_L represents the Langmuir adsorption isotherm constant (L/mg).

The Freundlich isotherm is given by Dudu et al. (2017), Eskhan et al. (2018), and Sharma et al. (2018a):

$$\log q_e = \log K_f + \left(\frac{1}{n} \right) \log C_e \quad (4)$$

where the parameters K_f and n are physical constants of the Freundlich isotherm and the value of n represents the intensity of the adsorption process and the mechanism of adsorption (physical or chemical). Thus, a value of n below unity indicates low or unfavorable adsorption intensity, suggesting physical adsorption. A value of n greater than unity indicates favorable adsorption intensity, implying chemical adsorption. For a value of n equal to unity, adsorption occurs linearly, with no predominance of one mechanism over the other (Sharma et al. 2018b).

In some cases, the adsorption phenomenon cannot be explained by the mentioned isotherms. Therefore, other models are used to describe the phenomenon. The following equation represents the Temkin isotherm (Jana et al. 2017, Priya et al. 2019):

$$q_e = \beta \log K_t + \beta \log C_e \quad (5)$$

where $\beta = RT/b$ is a constant representing the initial heat of adsorption and K_t an equilibrium constant. The Temkin isotherm suggests that the adsorption surface is a heterogeneous binding space with energy-containing junctions. These energies decrease linearly as electrostatic interactions between the adsorbent and adsorbate occur. On the other hand, β is a constant indicating the adsorbate's affinity for the adsorbent. Higher β values would signify a greater affinity for the active sites of the adsorbent (Jana et al. 2017, Priya et al. 2019).

Adsorption kinetics

Representative kinetics of dye adsorption processes are given by pseudo-first- and pseudo-second-order kinetics, whose governing equations are provided in equations 6 and 7, respectively (Jana et al. 2017, van Tran et al. 2018):

$$\frac{dq_t}{dt} = k_{p1}(q_e - q_t) \quad (6)$$

where q_t represents the mass of removed dye per mass of hydrogel (mg/g) during a time period t (min), and k_{p1} (L/min) is the rate constant of the pseudo-first-order kinetic model.

$$\frac{dq_t}{dt} = k_{p2}(q_e - q_t)^2 \quad (7)$$

where k_{p2} is the rate constant of the pseudo-second-order kinetic model.

Adsorption selectivity

Effluents generated by textile industries contain a variety of dyes in wastewater, often mixed and combined. This complexity poses challenges for efficient removal during adsorption processes. Consequently, assessing the selectivity of synthesized hydrogels against a group of dyes found in wastewater is crucial for predicting the performance of these polymers in real-world textile-industry scenarios (Jana et al. 2017).

The separation factor (α_Y^X) allows determining the interaction strength or preference of the adsorbent for a specific type of adsorbate. This comparative factor is calculated in pairs, as shown in the following equation (Jana et al. 2017).

$$\alpha_Y^X = \frac{q_X}{C_X} \frac{C_Y}{q_Y} \quad (8)$$

The decision rule states that when the factor α_Y^X is greater than one, adsorption by colorant X is

preferred; however, if the factor α_Y^X is less than one, selectivity will be inclined towards colorant Y .

Thermodynamics of adsorption

The feasibility and spontaneity of adsorption are determined through thermodynamic studies of the process, which establish whether the process is spontaneous or not, exothermic or endothermic, feasible or not. Additionally, thermodynamic parameters such as ΔH° (enthalpy change), ΔS° (entropy change) and ΔG° (Gibbs energy change) could be obtained (Sánchez et al. 2019, Vilela et al. 2019).

The standard free energy is obtained from (9):

$$\Delta G^\circ = -RT \ln K_{eq} \quad (9)$$

where the equilibrium constant K_{eq} is determined using (10):

$$K_{eq} = \frac{C_e}{C_i - C_e} \quad (10)$$

where C_i is the concentration of dye in the effluent and C_e the equilibrium concentration of dye in the effluent.

In addition, the standard entropy and enthalpy are calculated using the van 't Hoff equation:

$$\ln K_{eq} = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \quad (11)$$

where ΔS° represents the change in system entropy (J/mol.K), ΔH° is the change in system enthalpy (J/mol), R is the universal gas constant (J/mol.K), T is the temperature (K), and K_{eq} represents the equilibrium constant as a function of the system temperature.

The studies conducted for the removal of industrial dyes using hydrogels as an adsorption technology are diverse and extensive. One advantage of these polymers is their ability to undergo adsorption cycles without a significant drop in efficiency, making them sustainable and renewable materials for application in industrial textile effluent treatment. Another advantage is their potential for modification or coupling with other components (natural polymers, nanocomposites, clay materials, etc.) to enhance their dye-removal properties (Soleimani et al. 2018). **Table II** summarizes representative studies reported in the literature, including the most relevant data on adsorption performance, operating conditions, and hydrogel recyclability. Additionally, a discussion and analysis section is presented on the key parameters affecting adsorption efficiency: contact time, initial dye concentration, adsorbent dosage, ionic strength, temperature, and pH. The adsorption equilibrium is also discussed through isotherms, adsorption kinetics, and thermodynamics.

TABLE II. APPLICATION OF HYDROGELS IN THE REMOVAL OF INDUSTRIAL TEXTILE DYES - SUMMARY OF SELECTED STUDIES REPORTED IN LITERATURE.

Reference	Hydrogel	Colorant		Adsorption parameters				Recyclability	
		Adsorbent	Adsorbate	Charge	Maximum adsorption	pH	Isotherm	Kinetics	Thermodynamics
Dudu et al. (2017)	p(AMPS)- <i>Polyporus squamosus</i> (Huds.) Fr.	Toluidine blue (TB)	Cationic	40.9 mg/g (T = 25 °C, t = 24 h)	6.0	Freundlich (R ² = 0.9415)	Pseudo-first order (R ² = 0.9712)	-	-
Dudu et al. (2017)	p(APTMAcI)- <i>Polyporus squamosus</i> (Huds.) Fr.	Alizarin red (AR)	Anionic	31.3 mg/g (T = 25 °C, t = 24 h)	6.0	Freundlich (R ² = 0.8316)	Pseudo-second order (R ² = 0.9501)	-	-
Priya et al. (2019)	NaAla-Gel-cl-poly AAm/ZnS	Gallocyanine (G)	Anionic	36.7 mg/g (T = 25 °C, t = 24 h)	6.0	Freundlich (R ² = 0.9040)	Pseudo-second order (R ² = 0.9985)	-	-
Priya et al. (2019)	NaAla-Gel-cl-poly AAm/ZnS	Biebrich scarlet*	Anionic	97.37 % (T = 30 °C, t = 20 h)	5.0	Temkin (R ² = 0.9700)	Pseudo-first order (0.95 < R ² < 0.99)	Spontaneous and endothermic	5
Jana et al. (2017)	Poly(acrylamide-co-N-methylacrylamide)/KG (KG-6)	Crystal violet (CV)	Cationic	94.45 % (T = 30 °C, t = 16 h)	9.0	Freundlich (R ² = 0.9600)	Pseudo-second order (0.92 < R ² < 0.99)	Spontaneous and endothermic	5
Jana et al. (2017)	Poly(acrylamide-co-N-methylacrylamide)/KG (KG-6)	Methylene blue (MB)	Cationic	87.50 % (T = 25 °C, t ≈ 1 h)	8.0	Freundlich (R ² = 0.9986)	Pseudo-second order (R ² = 0.9999)	Spontaneous and exothermic	-
Yang et al. (2018)	Poly(acrylic acid-co-2-acrylamide-2-methyl-1-propanesulfonic acid)/kaolin (P(AA-co-AMPS)/KL)	Malachite green (MG)	Cationic	82.50 % (T = 25 °C, t ≈ 1 h)	6.0	Freundlich (R ² = 0.9993)	Pseudo-second order (R ² = 0.9999)	Spontaneous and exothermic	-
Yang et al. (2018)	Poly(acrylic acid-co-2-acrylamide-2-methyl-1-propanesulfonic acid)/kaolin (P(AA-co-AMPS)/KL)	Congo red (CR)	Anionic	85.00 % (T = 25 °C, t ≈ 1 h)	5.0	Freundlich (R ² = 0.9996)	Pseudo-second order (R ² = 0.9999)	Spontaneous and exothermic	-
Yang et al. (2018)	Poly(acrylic acid-co-2-acrylamide-2-methyl-1-propanesulfonic acid)/kaolin (P(AA-co-AMPS)/KL)	Methyl orange (MO)	Anionic	506 mg/g (T = 15 °C, t = 24 h)	7.0	Freundlich (R ² = 0.9736)	Pseudo-second order (R ² = 0.9889)	Spontaneous	-

Note: “-” indicates information not reported in the original study. T: Temperature. t: Time, R²: Coefficient of determination.

*Scarlet Biebrich = Scarlet Fuchsin = Acid Fuchsin

Own elaboration.

TABLE II. APPLICATION OF HYDROGELS IN THE REMOVAL OF INDUSTRIAL TEXTILE DYES - SUMMARY OF SELECTED STUDIES REPORTED IN LITERATURE.

Reference	Hydrogel	Colorant		Adsorption parameters				Recyclability cycles
		Adsorbate	Charge	Maximum adsorption	pH	Isotherm	Kinetics	
Peighambar-doust et al. (2020)	Carboxymethyl cellulose-g-polyacrylamide/montmorillonite (CMC-g-P(AAm)/MMT)	Malachite green (MG)	Cationic	94.914 % (T = 25-50 °C, t = 90 min.)	7.0	Langmuir (R ² = 0.9973)	Pseudo-second order (R ² = 0.9612)	-
Kıvanç et al. (2020)	Poly((methacryloylamino)propyl trimethylammonium chloride-co-vinylimidazole) (p(MAPTAC-co-VI))	Eriochrome black T (EBT)	Anionic	1818.2 mg/g (T = 20-60 °C, t = 2 h)	4.0-6.0	Langmuir (R ² = 0.9999)	Pseudo-second order (R ² = 0.9970)	-
		Methyl orange (MO)	Anionic	1449.3 mg/g (T = 20 °C-60 °C, t = 2 h)	4.0-6.0	Langmuir (R ² = 0.9999)	Pseudo-second order (R ² = 0.9999)	-
Dalalibera et al. (2020)	Polyacrylic acid-based (PAABH)	Methylene blue (MB)	Cationic	454.45 mg/g (T = 39.85-60 °C, t = 3500 min.)	8.0	Redlich-Peterson and Sips (R ² = 0.9980)	Pseudo-first order (R ² = 0.9928)	5
Rizzi et al. (2018)	Polyamidoamine-based hydrogel (PAA)	Direct red 83:1 (DR)	Anionic	98 % (T = 20-100 °C, t = 150 min.)	7.0	Temkin (R ² = 0.9999)	Pseudo-second order (R ² > 0.9999)	7
		Direct blue 78 (DB)	Anionic	93 % (T = 20-100 °C, t = 1 h)	7.0	Freundlich (R ² = 0.9996)	Pseudo-second order (R ² > 0.9999)	8
Elbedwehy and Atta (2020)	Polyacrylamide grafted Arabic gum (AG-g-PAM/PAA)	Methylene blue (MB)	Cationic	2 300 mg/g (T = 25 °C, t = 2 h)	7.0	Langmuir (R ² = 0.9950)	Pseudo-second order (R ² = 0.9997)	Greater than 3
Thamer et al. (2020)	Chitosan-grafted-polyacrylic acid/oxidized electropun carbon nanofiber (CT-g-PAA/O-ECNFs)	Methylene blue (MB)	Cationic	1095 mg/g (T = 25 °C, t = 24 h)	7.0	Freundlich (R ² = 0.9936)	Pseudo-second order (R ² = 0.9987)	Greater than 5
Wang et al. (2018)	Poly(vinyl alcohol)-sodium alginate-chitosan-montmorillonite nanosheets (MMTNS)	Methylene blue (MB)	Cationic	>90 % (T = 30 °C, t = 236 min.)	8.0	Redlich-Peterson and Sips (R ² ≈ 1.0000).	Pseudo-second order (R ² > 0.9900)	Greater than 5

Note: “-” indicates information not reported in the original study. T: Temperature. t: Time, R²: Coefficient of determination.

*Scarlet Biebrich = Scarlet Fuchsin = Acid Fuchsin

Own elaboration.

TABLE II. APPLICATION OF HYDROGELS IN THE REMOVAL OF INDUSTRIAL TEXTILE DYES - SUMMARY OF SELECTED STUDIES REPORTED IN LITERATURE.

Reference	Hydrogel	Colorant		Adsorption parameters				Recyclability cycles	
		Adsorbent	Adsorbate	Charge	Maximum adsorption	pH	Isotherm		Kinetics
Sharma et al. (2018b)	Gum arabic-cl-poly(acrylamide) nanohydrogel (GA-cl-poly(AAm))NHG	Crystal violet (CV)	Cationic	90.90 mg/g (T = 35 °C, t = 90 min.)	9.0	Langmuir (R ² = 0.9740)	Pseudo-first order (R ² = 0.9620)	Spontaneous and exothermic	6
Jana et al. (2019)	Katira gum-cl-poly(acrylic acid-co-N,N-dimethylacrylamide)/bentonite (KG-cl-poly(AA-co-DMA)/BT) (KGNCH)	Methylene blue (MB)	Cationic	165.28 mg/g (T = 50 °C, t = 80 min.)	8.0	Freundlich (R ² = 0.9950)	Pseudo-second order (R ² = 0.9950)	Spontaneous and endothermic	5
		Crystal violet (CV)	Cationic	158.73 mg/g (T = 50 °C, t = 80 min.)	7.5	Freundlich (R ² = 0.9954)	Pseudo-second order (R ² = 0.9940)	Spontaneous and endothermic	5
		Auramine O	Cationic	152.67 mg/g (T = 60 °C, t = 80 min.)	10.0	Freundlich (R ² = 0.9960)	Pseudo-second order (R ² = 0.9930)	Spontaneous and endothermic	5
		Crystal violet (CV)	Cationic	138.89 mg/g (T = 25-35 °C)	2.0-10.0	Langmuir (R ² = 0.9964)	Pseudo-second order (R ² = 0.9990)	Spontaneous and endothermic	5
Ren et al. (2021)	Gelatin hydrogel (GH)	Methyl orange (MO)	Anionic	72.63 mg/g (T = 25-35 °C)	2.0-10.0	Langmuir (R ² = 0.9941)	Pseudo-second order (R ² = 0.9993)	Spontaneous and endothermic	5
		Acid fuchsin* (AF)	Anionic	66.29 mg/g (T = 25-35 °C)	2.0-10.0	Langmuir (R ² = 0.9944)	Pseudo-second order (R ² = 0.9970)	Spontaneous and endothermic	5
		Congo red (CR)	Anionic	64.84 mg/g (T = 25-35 °C)	2.0-10.0	Langmuir (R ² = 0.9952)	Pseudo-second order (R ² = 0.9989)	Spontaneous and endothermic	5
		Malachite green (MG)	Cationic	64.64 mg/g (T = 25-35 °C)	2.0-10.0	Langmuir (R ² = 0.9980)	Pseudo-second order (R ² = 0.9994)	Spontaneous and endothermic	5
		Methylene blue (MB)	Cationic	60.32 mg/g (T = 25-35 °C)	2.0-10.0	Langmuir (R ² = 0.9908)	Pseudo-second order (R ² = 0.9938)	Spontaneous and endothermic	5

Note: “-” indicates information not reported in the original study. T: Temperature. t: Time, R²: Coefficient of determination.

*Scarlet Biebrich = Scarlet Fuchsin = Acid Fuchsin
Own elaboration.

TABLE II. APPLICATION OF HYDROGELS IN THE REMOVAL OF INDUSTRIAL TEXTILE DYES - SUMMARY OF SELECTED STUDIES REPORTED IN LITERATURE.

Reference	Hydrogel	Colorant		Adsorption parameters				Recyclability cycles		
		Adsorbent	Adsorbate	Charge	Maximum adsorption	pH	Isotherm		Kinetics	Thermodynamics
Ferfera-Harrar et al. (2021)	Chitosan-grafted-hydrolyzed polyacrylamide/montmorillonite		Basic red 46 (BR46)	Cationic	78 % (T=25 °C, t=3 h)	7.0	Langmuir (R ² = 0.9940)	Pseudo-second order (R ² = 0.9999)	Spontaneous and endothermic	4
Ren et al. (2021)	Graphene oxide - cellulose nanowhiskers nanocomposite (GO-CNW)	Methylene blue (MB)		Cationic	100 mg/g (t=20 min)	7.0	-	Pseudo-second order (R ² = 0.9953)	-	3
		Rhodamine B (RhB)		Cationic	90 % (t=40 min.)	7.0	-	Pseudo-second order (R ² = 0.9917)	-	3

Note: “-” indicates information not reported in the original study. T: Temperature. t: Time, R²: Coefficient of determination.

*Scarlet Biebrich = Scarlet Fuchsin = Acid Fuchsin
Own elaboration.

DISCUSSION

Contact time

The hydrogel's permanence in the solution is an important parameter for estimating the operational time of the hydrogel for dye removal. The adsorption process can be divided into three stages: (1) rapid adsorption with respect to contact time, due to the availability of active sites on the hydrogel; (2) gradual and slow adsorption, owing to the decrease in available active sites for dye adsorption; and (3) minimal and equilibrium adsorption, resulting from the saturation of active sites available for dye adsorption. Additionally, the adsorption mechanism occurs

through molecular transport and intermolecular diffusion. The initial step results from the entrainment of molecules present in the solution toward the hydrogel surface (adsorbent). The second step takes place due to interactions between positive and negative charges between the hydrogel and the dye; diffusion occurs through the hydrogel pores, from its surface to active centers (Dalalibera et al. 2020).

The main forces involved in adsorption are Van der Waals forces, hydrogen bonding (H-bonding), and electrostatic interactions.

The adsorption percentage increased with contact time, as shown in **Figure 1a**. Priya et al. (2019) employed concentrations of 20 and 10 mg/L for

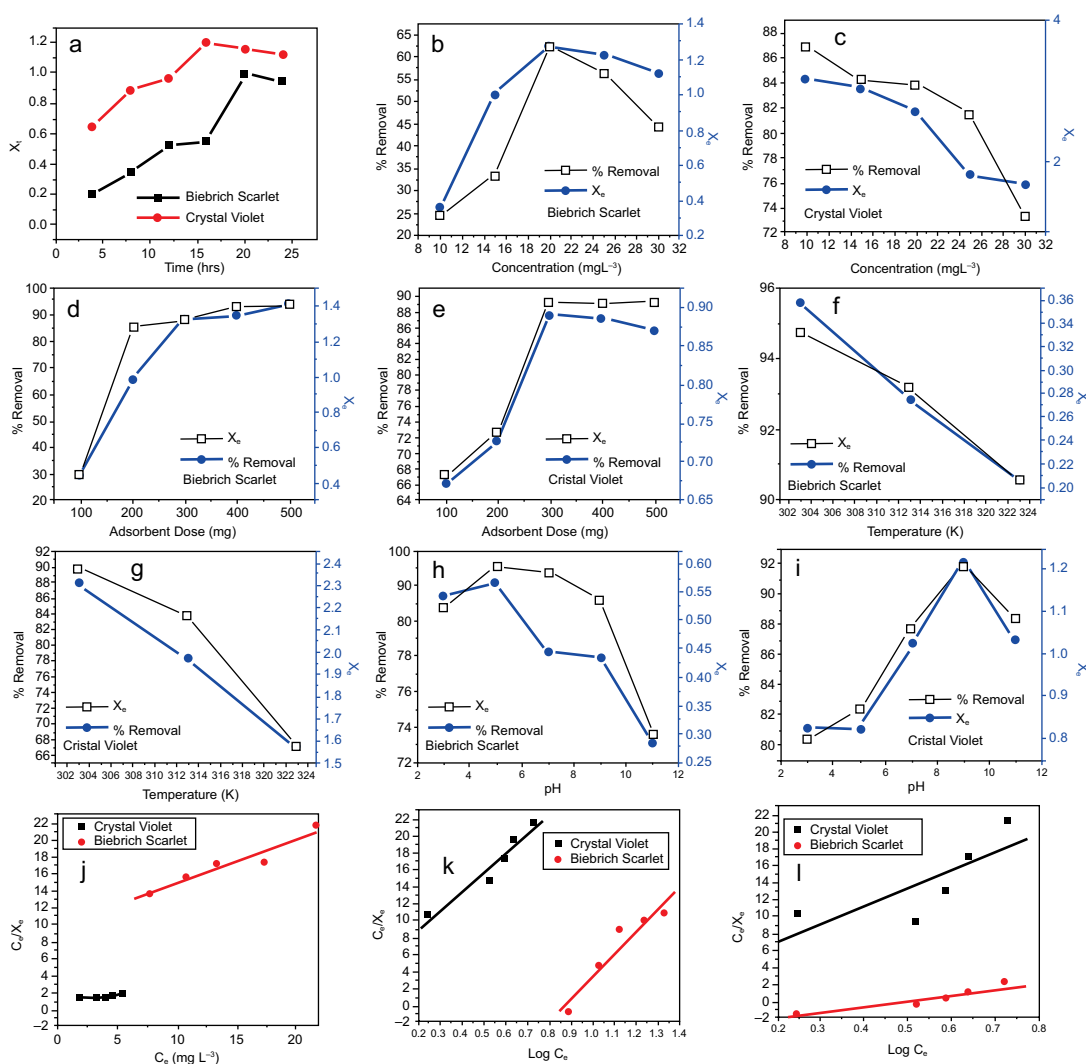


Fig. 1. Effect of adsorption parameters on removal efficiency: (a) removal time, (b-c) initial dye concentration, (d-e) adsorbent dosage, (f-g) temperature, (h-i) pH, (j) Langmuir isotherm, (k) Freundlich isotherm, and (l) Temkin isotherm (Priya et al. 2019). (X_t : amount of dye adsorbed at time t per unit mass of adsorbent; X_e : amount of dye adsorbed at equilibrium per unit mass of adsorbent).

Biebrich scarlet and crystal violet dyes, respectively. The results showed adsorption levels of 97.37 and 94.45% within 20 and 16 h for Biebrich scarlet and crystal violet, respectively.

Initial dye concentration

The interaction between hydrogels and dyes occurs through their respective ions; however, initial dye concentration influences whether this interaction occurs. At very low concentrations, the hydrogel surface ions cannot interact with the dye ions because of the distance or dispersion between them. As dye concentration increases, the interaction is enhanced, creating active sites occupied by dye ions. However, the number of active sites is limited (at a fixed initial hydrogel concentration), and their saturation depends on the initial dye concentration (Dalalibera et al. 2020, Ren et al. 2021).

Dudu et al. (2017) conducted a dye removal study involving TB (toluidine blue), AR (alizarin red), and MG (malachite green), where they varied the initial dye concentration from 10 to 100 mg/L (hydrogel dose = 1 g/L, contact time = 24 h), observing its effect on removal efficiency. Additionally, studies by Priya et al. (2019) achieved a maximum adsorption percentage of 97.37 and 94.45% using semi-IPN NaAla-Gel-cl-polyAAM hydrogels for Biebrich scarlet and crystal violet dyes, respectively, at initial concentrations of 20 and 10 mg/L (**Fig. 1b-c**). Furthermore, Bento et al. (2020) investigated the removal of MB (methylene blue), MG, and CR (congo red), varying the initial dye concentrations from 20 to 80 mg/L. They reported removal percentages of 87.46–82.55% for MB, 81.65–77.05% for MG, and 71.39–76.34% for CR, indicating that adsorption efficiency varies by dye type and its interaction with the hydrogel matrix (Jana et al. 2017).

Dose of adsorbent

The active sites of hydrogels function to capture ions through physical bonds. These vacancies become saturated as adsorption progresses, reaching an equilibrium state in which the hydrogel is unable to continue the adsorption process due to a lack of available active sites. The mass requirement of the adsorbent is directly proportional to the availability of active sites; therefore, a greater amount of hydrogel results in higher removal of dye from the medium. However, it is necessary to quantify the required amount of hydrogel to avoid overutilization of material and yield loss (Jana et al. 2017).

Priya et al. (2019) determined that the optimal dosage of semi-IPN NaAla-Gel-cl-polyAAM hydrogels

for the removal of Biebrich scarlet and crystal violet was 16.67 and 10 mg/mL, achieving removal efficiencies of 93.43 and 89.1%, respectively; see **Figure 1d-e** (Jana et al. 2017), which shows that increasing the hydrogel dosage improves the dye removal efficiency up to the optimal point, after which no significant enhancement is observed, suggesting that adsorption sites become saturated at higher dosages. These results highlight the importance of optimizing hydrogel concentration to maximize dye removal while avoiding excessive material usage.

Likewise, Priya et al. (2019) reported that removal percentage increased as adsorbent dosage increased. This is attributed to the greater availability of active sites in the solution for dye removal. However, once the removal limit (equilibrium) is reached, further addition of adsorbent will not increase the efficiency (Jana et al. 2017).

Ionic strength

In most textile industries, chemical pre-treatments are carried out as a means of conditioning or cleaning materials such as garments, leather, cotton, etc. These treatments release by-products or residues with various ions into the solution, which accumulate and mix with dyes for removal from the process as effluents. These ionic compounds or ions carry charges and dissolve in the medium (effluent) along with the dyes. Ionic strength between the hydrogel, dye, and dissolved ions can positively or negatively affect the dye-removal efficiency. For instance, if a cationic dye is adsorbed in a medium containing Na⁺ ions using an anionic adsorbent, the dye ions will compete with the solution's ions for the active sites on the adsorbent. Conversely, if the same process is carried out in a medium composed of Cl⁻ ions, no competition will occur because the medium and the hydrogel surface possess the same surface charge.

Particle size also influences adsorption, as it has an inversely proportional relationship with removal efficiency. In simpler terms, smaller particle sizes result in higher adsorption. Dyes have complex structures with long, heavy chains; conversely, the ionic charges dissolved in the solution are simple and small. Consequently, hydrogels prefer the dissolved ions. This means active sites will be mostly occupied by ions dissolved in the medium, leading to a significant decrease in dye adsorption efficiency.

Studies by Jana et al. (2017) showed a decrease in the removal efficiency of dyes MB, MG, and CR as the concentration of NaNO₃ increased, reaching values of 78, 72, and 67% at a 1 M concentration of NaNO₃.

Temperature

One disadvantage of working with polymers is their sensitivity to temperature changes. Hydrogels are three-dimensional materials whose polymeric structure is formed by networks or meshes that carry charges on their hydrophilic surface, capable of adsorbing ions with opposite charges. Changes in temperature within a solution can influence the adsorption efficiency, either positively (increasing adsorption) or negatively (decreasing adsorption).

Priya et al. (2019) investigated the effect of temperature on the removal of Biebrich scarlet and crystal violet dyes at 30, 40, and 50 °C. They determined that the optimal removal temperature is 30 °C, as at higher temperatures, the adsorption efficiency began to decrease (**Fig. 1 f-g**). The removal percentages obtained at this temperature were 94.76 and 89.85% for Biebrich scarlet and crystal violet dyes, respectively. However, this decrease in the adsorption percentage does not drop below 90% even when the operating temperature reaches 50 °C.

The behavior of hydrogels in response to temperature variations depends primarily on the type of polymer used in their synthesis. There are hydrogels that, with increasing temperature, experience contractions in their binding networks, causing surface ions to repel interactions with the fluid and leading to a decrease in adsorption. On the other hand, some hydrogels show an increased adsorption percentage as the solution temperature rises. This is due to relaxation in the binding chains and an increase in the stretching capacity of the hydrogel's mesh structure.

Figure 1f-g shows that the dyes MB, MG, and CR experienced an increase in molecular movement at elevated temperatures (50 and 60 °C), which provided them enough energy to facilitate interaction with the functional groups of the hydrogels, leading to enhanced removal efficiency.

pH

The pH behavior is similar to that of ionic strength, meaning it can disrupt dye removal efficiency due to variations in ionic charges in the medium. However, these pH variations can result from the process itself or from personnel intervention as a pre-treatment measure before adsorption. Analyzing pH variation in relation to the percentage of dye removal aims to study and determine the conditions favorable for maximum dye removal from the medium.

If the working solution has a pH below 7, cationic dyes will be scarcely removed from the medium by hydrogels due to the existing competition between the medium's H^+ ions and the adsorbate's (H^+) ions

to occupy the active sites of the hydrogel (OH^-). In a medium where H^+ ion concentration is significant and the goal is to remove a cationic dye (H^+), the active centers of hydrogels, which have an anionic (OH^-) surface charge, can potentially be occupied by H^+ ions from both the medium and the dye. These ions compete to occupy vacancies within the hydrogel's active sites, quickly saturating them without achieving complete dye removal. An example of this can be seen in the study by Dudu et al. (2017), where the adsorption of AR (OH^-) using p(APTMACl)-*Polyporus squamosus* (Huds.) Fr. at pH 6.0 resulted in an adsorption percentage of 43.6%. Similarly, Jana et al. (2017) observed that the removal of MB (H^+) and MG (H^+) using KG-6 hydrogels at pH levels below 7 negatively affected the efficiency of dye removal. Conversely, in a basic solution, the adsorption of cationic dyes is favored because hydrogels, being anionic (OH^-), remain unaffected, and their active centers are not saturated by the medium's ions. As a result, electrostatic interaction and the adsorption process are positively influenced.

Likewise, Munagapati et al. (2023) synthesized a novel Fe_3O_4 -GLP@CAB material for MB removal. Batch experiments showed a maximum removal efficiency of 95.2% at pH 10.0, with Langmuir isotherm data showing excellent agreement. Kinetic analysis suggested physisorption predominated, with stable performance over five regeneration cycles. This material proves highly recyclable and effective for MB dye removal from textile industrial effluents.

Dudu et al. (2017) concluded that for TB (H^+) removal using p(AMPS)-*Polyporus squamosus* (Huds.) Fr., the maximum removal efficiency reached 73.2% at pH 8. For the removal of G dye (OH^-) through the p(APTMACl)-*Polyporus squamosus* (Huds.) Fr. hydrogel, a maximum removal percentage of 69.4% was achieved at a pH of 6.0. In contrast, Jana et al. (2017) showed that Biebrich scarlet (OH^-) and crystal violet (H^+) dyes reached maximum removal percentages of 95.26% and 91.68%, respectively, at pH 5.0 and 9.0 (**Fig. 1 h-i**). Similarly, removal of CR (OH^-) in basic media (pH > 5.5) showed low removal percentages below 65%. However, this type of medium was ideal for the removal of MB (H^+) and MG (H^+).

The nature of the medium (acidic or basic) is determined through the parameter pH_{zpc} (pH of zero-point charge), which indicates the influence of changes in H^+ or OH^- ions in the medium. Thus, at pH values < pH_{zpc} , the medium carries a positive charge configuration, favoring the adsorption of anionic dyes due to increased electrostatic interaction. Conversely, at pH values > pH_{zpc} , the medium carries a negative

charge configuration, favoring the adsorption of cationic dyes (Kıvanç et al. 2020).

Adsorption isotherms

The most commonly used isotherms to determine adsorption equilibrium are the Langmuir and Freundlich isotherms, which offer a physicochemical approach for modeling experimental equilibrium data.

The results of Dudu et al. (2017) concluded that the equilibrium adsorption values of TB, AR, and MG colorants using p(AMPS)-*Polyporus squamosus* (Huds.) Fr. and p(APTMACl)-*Polyporus squamosus* (Huds.) Fr. hydrogels were well represented and fitted by the Freundlich isotherms. The parameter n was greater than unity for all three adsorption processes, indicating favorable adsorption of TB, AR, and MG.

On the other hand, Priya et al. (2019) presented adsorption isotherm results for Biebrich scarlet and crystal violet colorants (Fig. 1j-l). Among the tested models, the Freundlich isotherm (Fig. 1k) showed n values of 0.81 and 0.97 for Biebrich scarlet and crystal violet, respectively, suggesting that the adsorption process was dominated by chemical interactions (chemisorption). The Langmuir (Fig. 1j) and Temkin (Fig. 1l) models were also evaluated, providing complementary insights into monolayer adsorption capacity and adsorption energy distribution.

Adsorption kinetics

Adsorption kinetics allow us to determine the rate or amount of dye adsorbed per unit of time. This enables the study of the adsorption process over the time required to remove the dye from the medium. The curves from experimental dye-removal data exhibit standard behavior. Initially, the removal rate is high due to the abundance of active sites. However, as the adsorption time increases, this rate gradually decreases until it becomes constant, indicating saturation of active sites on the hydrogel. The kinetic models commonly applied to dye adsorption processes are pseudo-first-order and pseudo-second-order.

Dudu et al. (2017) reported that the hydrogel p(AMPS)-*Polyporus squamosus* (Huds.) Fr. removed 44% of TB after 24 h of contact time. Similarly, for AR and MG, removal efficiencies of 65.2 and 77.3% were achieved after 420 and 480 min, respectively, using the hydrogel p(APTMACl)-*Polyporus squamosus* (Huds.) Fr. These results indicate that dye uptake increases progressively with contact time until approaching equilibrium, which is characteristic of adsorption systems where the rate of adsorption is initially rapid due to the availability of active sites and gradually decreases as these sites become occupied.

Although the study mainly reports removal efficiencies rather than kinetic constants, the described time-dependent adsorption behavior is consistent with adsorption systems commonly well fitted by the pseudo-second-order kinetic model, suggesting that the adsorption process is likely governed by chemisorption mechanisms involving interactions between dye molecules and the functional groups present in the hydrogel network.

Adsorption thermodynamics

The viability, spontaneity, and type of adsorption are determined through thermodynamic analysis. The important variables for the analysis are given by equations 9, 10, and 11. The enthalpy change (ΔH°) indicates whether the adsorption process releases energy (exothermic) or requires energy (endothermic), while the entropy change (ΔS°) reflects the variation in randomness at the solid-solution interface. The Gibbs free energy change (ΔG°) is used to evaluate the feasibility and spontaneity of the adsorption process.

Priya et al. (2019) conducted thermodynamic studies on dye adsorption, reporting ΔS° and ΔH° values of 0.019 and $-10.621 \text{ kJ mol}^{-1}$ for Biebrich scarlet dye, and 0.0023 and $-11.634 \text{ kJ mol}^{-1}$ for crystal violet dye, respectively. The negative ΔH° values indicate that the adsorption process is exothermic. In addition, the positive ΔS° values suggest an increase in randomness at the adsorbent-solution interface during adsorption. Furthermore, at temperatures of 303, 313, and 323 K, the ΔG° values were -44.03 , -41.65 , and $-39.26 \text{ kJ mol}^{-1}$ for crystal violet, and -48.53 , -46.63 , and $-44.73 \text{ kJ mol}^{-1}$ for Biebrich scarlet dye, respectively. The negative ΔG° values confirm that the adsorption process is thermodynamically feasible and spontaneous within the studied temperature range.

Cycles of reuse and regeneration

Among the characteristics of hydrogels, two notable features are: (1) the elastic deformation and recovery of the network or mesh and (2) the electrostatic attraction of the hydrogel's surface charge to the dye ions. The first feature enables the hydrogel to return to its initial state before the adsorption process, providing it with repeatability and renewability capacity. This means that hydrogels can be reused in the adsorption process multiple times without significantly affecting their properties and effectiveness. Regarding the second feature, electrostatic attraction is a weak or superficial force. Through standard treatments, hydrogels can be regenerated, releasing active sites, and allowing the hydrogel to adsorb dyes from the medium again while maintaining its efficiency.

Priya et al. (2019) conducted a washing process using NaOH 0.1 M and HCl 0.1 M for the desorption of Biebrich scarlet and crystal violet, respectively. After the regeneration process, these dyes were reused as adsorbents for dye removal with the same properties and effectiveness. However, the extent to which the polymer can be regenerated and reused is not unlimited, as shown in **table II**, where the reported studies generally indicate a maximum of five reuse cycles. The cycles of reuse depend on the percentage of removal obtained after each regeneration and the acceptable level of tolerance.

On the other hand, Priya et al. (2019) also conducted recyclability studies for NaAla-Gel-cl-polyAAm/ZnS hydrogels and determined that the hydrogel's lifespan included up to five reuse cycles before a decrease in adsorption efficiency was observed. This decrease was attributed to the denaturation of the polymer matrices of the hydrogel and deactivation of active sites.

Selectivity

Jana et al. (2017) studied the removal of ternary dye mixtures of MB, MG, and CR using KG-6 hydrogels. Selectivity analysis (see equation 8) was conducted for the MB/MG, CR/MB, and CR/MG dye pairs. The results determined that KG-6 hydrogels exhibit higher affinity towards MB compared to MG. Moreover, it was found that the selectivity for MG is higher than for CR. Ultimately, KG-6 hydrogel demonstrates greater removal affinity for MB than for CR, thus establishing the selectivity hierarchy as follows: MB > MG > CR.

To understand selectivity, two factors must be considered: the surface charges of the dyes and their particle size (spatial structure). The forces of attraction between ionic surfaces and dyes represent the affinity strength between both materials; dyes with higher attraction forces are more likely to be adsorbed. Furthermore, particle size influences the occupancy of active sites within the hydrogel's structure, so smaller particle sizes positively affect the removal or adsorption process.

CONCLUSIONS

Global growth in recent years has driven economic and technological advancements in the textile industry. However, this progress has also generated significant amounts of contaminants and industrial textile waste, particularly industrial effluents with high concentrations of textile dyes that pollute water

bodies such as lakes, rivers, and beaches. This has prompted the scientific community to seek alternative technologies to decontaminate these effluents before discharge. Sustainable and renewable technologies that enable continuous implementation in effluent treatment have garnered significant attention in this pursuit.

This study compiles recent advances in applying sustainable smart hydrogels to treat textile industrial effluents under different operating conditions. The synthesis table summarizes key adsorption parameters reported in the literature, including equilibrium behavior (isotherms), adsorption kinetics, thermodynamic analysis, and the recyclability cycles of hydrogels. These parameters provide relevant information for evaluating the potential application of hydrogels in the removal of textile dyes from industrial effluents.

Nomenclature

q_e	Adsorption capacity at equilibrium
C_0	Dye concentration at initial time
C_e	Dye concentration at equilibrium time
V	Volume of dye solution
W	Weight of adsorbent
$q_{\max}$	Theoretical maximum adsorption capacity
K_L	Langmuir adsorption isotherm constant
K_f	Freundlich adsorption isotherm constant
n	Freundlich isotherm constant
β	Initial heat of adsorption
K_t	Temkin adsorption isotherm constant
b	Temkin constant related to the heat of adsorption
q_t	Adsorption capacity at time t
k_{p1}	Rate constant of the pseudo-first-order kinetic
t	Time
k_{p2}	Rate constant of the pseudo-second-order kinetic
α_Y^X	Separation factor of a single dye in comparison of two systems
q_X	Adsorption capacity of X dye
q_Y	Adsorption capacity of Y dye
C_X	Concentration of dye X
C_Y	Concentration of dye Y
ΔG°	Gibbs energy change
R	Universal gas constant
T	Temperature
K_{eq}	Equilibrium constant
C_i	Dye concentration in the effluent
ΔS°	Change in system entropy
ΔH°	Change in system enthalpy
TB	Toluidine Blue (cationic dye)
AR	Alizarin Red (anionic dye)

MG Malachite Green (cationic dye)
 MB Methylene Blue
 CR Congo Red
 PH_{zpc} PH of zero-point charge

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