

# Waste-derived hydrogels for pharmaceutical effluent treatment: a review of synthesis, modification, applications, and future perspectives

Hidrogéis derivados de resíduos para o tratamento de efluentes farmacêuticos: uma revisão sobre síntese, modificações, aplicações e perspectivas futuras

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## ABSTRACT

The presence of pharmaceuticals in wastewater and surface water is a growing environmental concern due to their persistence and potential ecotoxicological impacts. Conventional wastewater treatment methods often fail to effectively remove these contaminants, prompting research into innovative remediation technologies. In this context, hydrogels synthesized from agricultural and industrial residues have emerged as promising materials for the selective adsorption of pharmaceuticals. This systematic review addresses recent advancements in the synthesis and modification of waste-derived hydrogels, emphasizing their physicochemical properties, adsorption mechanisms, and removal efficiencies. The analysis reveals that, despite the abundance of research on the utilization of residual biomass, targeted applications for pharmaceutical removal are still limited. In terms of performance, waste-based hydrogels demonstrate high adsorption capacities. The predominant mechanisms are electrostatic interactions,  $\pi$ - $\pi$  interactions, hydrogen bonding, and electron donor-acceptor (EDA) interactions. In conclusion, waste valorization significantly contributes to mitigating pharmaceutical pollution.

**Keywords:** waste-derived hydrogels; adsorption mechanisms; circular economy; adsorption of pharmaceuticals.

## RESUMO

A presença de produtos farmacêuticos em águas residuais e superficiais é uma crescente preocupação ambiental, em razão da sua persistência e dos potenciais impactos ecotoxicológicos. Os métodos convencionais de tratamento de águas residuais frequentemente falham na remoção eficaz desses contaminantes, o que impulsiona a pesquisa de tecnologias inovadoras para remediação. Nesse contexto, hidrogéis sintetizados de resíduos agrícolas e industriais têm surgido como materiais promissores para a adsorção seletiva de produtos farmacêuticos. Esta revisão sistemática aborda os avanços recentes na síntese e modificação de hidrogéis derivados de resíduos, enfatizando suas propriedades físico-químicas, mecanismos de adsorção e eficiências de remoção. Esta análise revelou que, apesar da abundância de pesquisas sobre a utilização de biomassa residual, as aplicações direcionadas à remoção de produtos farmacêuticos ainda são limitadas. Em termos de desempenho, os hidrogéis à base de resíduos demonstram altas capacidades de adsorção. Os mecanismos predominantes são interações eletrostáticas, interações  $\pi$ - $\pi$ , ligações de hidrogênio e interações doador-aceitador de elétrons (EDA). Assim, conclui-se que a valorização de resíduos contribui significativamente para a mitigação da poluição farmacêutica.

**Palavras-chave:** hidrogéis derivados de resíduos; mecanismos de adsorção; economia circular; adsorção de produtos farmacêuticos.

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Conflicts of interest: the authors declare no conflicts of interest.

Funding: Coordenação de Aperfeiçoamento de Pessoal de Nível Superior, Conselho Nacional de Desenvolvimento Científico Tecnológico (Grant # 305910/2023-0) and Financiadora de Estudos e Projetos (Grant # 01.23.0211.01).

Received on: 01/12/2026. Accepted on: 03/11/2026.

<https://doi.org/10.5327/Z2176-94782943>



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## Introduction

The escalating occurrence of pharmaceuticals in aquatic effluents has generated significant environmental and public health concerns (Morillas-España et al., 2025). The indiscriminate use of medications, coupled with the lack of appropriate effluent disposal, results in the discharge of these compounds into the environment, adversely impacting aquatic ecosystems and potentially human health (Zeng et al., 2024). Many pharmaceuticals are not completely metabolized within animal organisms, and their inadequate removal through conventional wastewater treatment plants can result in sublethal concentrations of pharmacological substances in receiving water bodies. This scenario underscores the pressing urgency to develop innovative and effective remediation technologies for the removal of these contaminants, particularly from wastewater matrices (Ghosh et al., 2023).

Furthermore, the identification of effective treatment methodologies capable of addressing the diversity of pharmacological compounds present in various effluent streams remains one of the primary challenges for environmental managers (Ma et al., 2020). Removal processes requires high efficiency in addition to being cost-effective and sustainable. Advancements in contaminant adsorption technologies, such as hydrogels, have emerged as a promising alternative due to their capacity for selective and efficient pharmaceutical removal (Li et al., 2024).

Hydrogels are three-dimensional materials composed of polymeric networks that can retain substantial quantities of water within their structures (Maitra and Shukla, 2014). This capacity for water absorption is a primary characteristic of these materials, allowing them to expand or contract (swelling/deswelling) based on their specific composition and response to environmental stimuli. Physical properties, such as elasticity and high hydrophilicity, enable hydrogels to be highly efficient in the adsorption of various substances, including organic contaminants such as pharmaceuticals. These attributes make hydrogels ideal materials for the treatment of contaminated wastewater (Maitra and Shukla, 2014).

In addition to their inherent physical attributes, hydrogels can be chemically modified to enhance their affinity for specific compounds, such as pharmaceuticals (Mittal et al., 2023). Surface functionalization can significantly improve their removal capacity for targeted pollutants, therefore acting as a versatile option across diverse remediation scenarios (Wang, L. et al., 2025). This versatility, coupled with their inherent tailorability, is a primary reason why hydrogels are regarded as an innovative solution for environmental challenges involving pharmaceutical effluents (Rezvani-Ghalhari et al., 2024).

The use of industrial and agricultural residues in hydrogel synthesis not only provides a sustainable alternative for the production of these materials but also facilitates the mitigation of environmental impacts resulting from the inappropriate disposal of such waste (Zhou et al., 2025). The use of residual biomass, including fruit peels, food waste, or agro-industrial residues, can serve as effective raw materi-

als for the development of hydrogels with physicochemical properties comparable to synthetic counterparts, while offering the distinct benefits of being derived from renewable and cost-effective resources (Rezvani-Ghalhari et al., 2024).

This practice is advantageous from both environmental and economic perspectives, as it minimizes the reliance on petrochemical resources and mitigates the volume of waste accumulated in landfills and the natural environment, thereby advancing the Sustainable Development Goals (SDGs). Furthermore, the valorization of agricultural and industrial residues for the development of functional, high-value-added materials, such as hydrogels, has become a prominent focus of sustainability research (B. Tang et al., 2025). Consequently, beyond enhancing the efficacy of pharmaceutical remediation in effluents, this approach promotes a robust circular economy, wherein waste is transformed into high-utility novel materials (X. Tang et al., 2025).

Although research involving sustainable hydrogels has expanded significantly in recent years, a substantial portion of these studies focuses on the removal of contaminants such as dyes, heavy metals, and nutrients (Kaur et al., 2025; Li et al., 2026), whereas applications specifically targeted toward the removal of pharmaceuticals remain relatively underexploited. In the case of hydrogels synthesized from agro-industrial residues, this research gap becomes even more pronounced. Current literature exhibits a predominance of synthetic materials or hydrogels subjected to extensive chemical modifications aimed at enhancing selectivity and adsorptive performance. In contrast, the feasibility, efficiency, and underlying adsorption mechanisms of hydrogels derived from renewable residues for pharmaceutical remediation continue to be addressed in a limited and insufficiently systematized manner (Du et al., 2024; Wang, L. et al., 2025).

Consequently, it is essential to advance research regarding the synthesis, functionalization, and performance of these sustainable hydrogels to ensure their efficacy and expand their implementation as a promising strategy for effluent remediation. The objective of this work is to review and discuss the applicability of hydrogels, particularly those synthesized from agro-industrial residues, for the adsorption of pharmaceuticals in aquatic effluents. This review also addresses chemical modification techniques for hydrogels and the outcomes reported in empirical pharmaceutical removal tests.

## Methodology

The methodology adopted herein consists of a systematic literature review. Recent studies addressing the synthesis, characterization, and physicochemical properties of hydrogels were analyzed, evaluating their adsorption capacities for contaminants, as well as the environmental and economic impacts (socioeconomic impacts) resulting from the utilization of agro-industrial residues as a primary feedstock for material production. The bibliographic search was conducted across the Web of Science and Scopus databases, employing the following keywords and their Boolean combinations (and/or): “Hydrogel”, “Phar-

maceuticals wastewater treatment”, “Adsorption of pharmaceuticals”, “Waste-derived hydrogel”, “Sustainable hydrogels”. The scope has been restricted to articles published over the last ten years (2017–2026), with a specific emphasis on relevant experimental studies within the thematic area.

Complementing the qualitative assessment, a bibliometric analysis was conducted using the R software (bibliometrix package) to discern emerging trends and map collaboration networks among authors and institutions, as well as to evaluate the trajectory of scientific output within this field. The inclusion criteria for the selected articles prioritized relevance, scope, and methodological rigor. Conversely, the exclusion criteria targeted duplicate entries, papers lacking full-text accessibility, or those that did not align with the predefined goals of this review.

## Results and Discussion

### Raw materials origin for hydrogel fabrication

The natural raw materials for hydrogel synthesis are predominantly based on renewable polysaccharides and proteins, which are highly valued for their biodegradability and low toxicity. Cellulose, the most abundant natural polymer, is primarily extracted from the cell walls of higher plants, such as trees and cotton, and is also recovered from agricultural residues including rice husks, banana peels, and sugarcane bagasse (Mittal et al., 2023; Verma et al., 2025). Another plant-based polysaccharide of significance is starch, obtained from tubers and grains such as potatoes, corn, and wheat (Hossain et al., 2023; Khoo et al., 2023). Furthermore, natural gums extracted from plant exudates or seeds, such as gum arabic, gum ghatti, and gum tragacanth, provide a polymeric network rich in hydroxyl and carboxyl groups, which are essential for water affinity and swelling performance (Mittal et al., 2023; Du et al., 2024).

In contrast, the marine environment and the animal kingdom provide biopolymers with unique electrostatic properties, such as alginate and carrageenan, extracted from brown and red algae (seaweed), respectively, through alkaline extraction processes (Ahmed et al., 2024; Ijaz et al., 2024). Chitin and its deacetylated derivative, chitosan, are cationic polymers obtained from the exoskeleton of crustaceans, such as shrimp and crabs, as well as the cell walls of fungi and insects (Khumalo et al., 2024; Yang et al., 2025). Regarding proteins, collagen and gelatin are isolated from mammalian connective tissues, bones, and skins, whereas the chondroitin sulfate is commonly extracted from vertebrate cartilage, offering active sites for complex interactions within hydrogel networks (Hossain et al., 2023; Liu et al., 2025).

The use of industrial residues and biological by-products represents a sustainable frontier in obtaining these raw materials. Lignin, the second most abundant organic polymer on Earth, is extracted as a byproduct of pulping processes in the paper industry, serving as a primary renewable source of aromatic structures (Mondal et al., 2021; Raj et al., 2025). Urban waste, such as discarded newspaper, is also pro-

cessed for the recovery of low-cost cellulose fibers (Rezvani-Ghalhari et al., 2024). Additionally, microbial-derived polymers, such as xanthan gum (produced by the bacterium *Xanthomonas campestris*) and gellan gum, are generated via fermentation, enabling the production of hydrogels with precise control over purity and structural properties (Mittal et al., 2023; Alali et al., 2026).

From an integrated perspective, the diversity of natural sources, including plant-based, marine, animal-based, microbial, and industrial waste-derived origins, significantly expands the spectrum of raw materials available for hydrogel synthesis. This wide range of options not only ensures tunable physicochemical and functional properties but also reinforces the alignment of these materials with the principles of sustainability, the circular bioeconomy, and byproduct valorization.

### Synthesis and modification of waste-derived hydrogels

The synthesis of hydrogels from waste is an innovative practice with the potential to advance sustainability in the use of materials and natural resources (Yang et al., 2025). The first step in the synthesis of waste-derived hydrogels involves the preparation and purification of the starting materials (Hossain et al., 2023). The purification process comprises stages such as the removal of soluble impurities and drying. Organic residues contain water- or acid-soluble components that must be eliminated, including sugars, fatty acids, or proteins. Several techniques may be employed, such as washing with diluted acids or the use of ethanol, followed by oven or freeze-drying (lyophilization), which ensures high material purity (Verma et al., 2025).

In the study by Prasannamedha et al. (2022), sugarcane bagasse was converted into magnetic hydrogel microspheres (SACFe) through a two-step process. First, the biomass was turned into magnetic carbon via hydrothermal carbonization followed by thermal activation. For the microsphere fabrication, sodium alginate was dissolved in water at 60°C, where the magnetic carbon (previously sonicated for uniform dispersion) was incorporated until complete homogenization. The viscous suspension was then added dropwise into a calcium carbonate solution, which acted as an ionic cross-linking agent, resulting in the instantaneous solidification of beads with a 2 mm diameter (Prasannamedha et al., 2022).

The valorization of urban waste, such as newspapers, for cellulose extraction represents a central strategy for sustainability. Rezvani-Ghalhari et al. (2024) describe an extraction process that employs NaOH and sodium bicarbonate solutions to remove paper additives, such as ink and oil, from newspaper fibers (which consist of approximately 68% cellulose). The adsorbent synthesis is conducted via the sol-gel method, wherein the extracted cellulose is blended with polyvinyl alcohol (PVA) and mixed metal oxides (MMO). The final structure is consolidated by encapsulation, through the injection of the mixture into a linker solution containing calcium chloride and boric acid, followed by lyophilization (freeze-drying) generating a highly porous and fibrous architecture (Rezvani-Ghalhari et al., 2024).

An advanced biotechnological approach involves the development of self-reinforcing “living hydrogels” from industrial effluents. X. Tang et al. (2025) developed an in-situ synthesis strategy by combining the *Rhizopus oryzae* fungus with TEMPO-mediated oxidation-derived cellulose nanofibers (TOCN). The key mechanism relies on fungus fermentation, which produces fumaric acid. The liberated H<sup>+</sup> ions then neutralize the negative charges on the carboxyl groups of the nanofibers, facilitating the overlapping of the TOCN fibers via hydrogen bonding. This process immobilizes the microorganism within the polymeric network, leveraging mycelial proliferation to enhance the elastic modulus of the material (*G'*) during wastewater treatment (X. Tang et al., 2025).

In contrast, the modification of natural gums via radiation technologies offers a clean and rapid alternative to conventional methods. Du et al. (2024) explored the functionalization of gum tragacanth with ionic liquids using electron beam irradiation. In this synthesis, the blend of gum and ionic liquids was subjected to radiation doses (up to 80 kGy), which induced the formation of free radicals on both the gum and the functional molecules, triggering simultaneous graft polymerization and cross-linking. This technique eliminates the need for toxic chemical initiators and allows for precise control over the gel fraction and the swelling ratio of the final hydrogel (Du et al., 2024).

Finally, a notable example is the integration of hybrid biopolymers, such as cyclodextrin and carboxymethyl chitosan, which enables the development of multifunctional networks via free radical polymerization. Zhang et al. (2026) described the construction of the ADS-Gel-1 hydrogel utilizing  $\alpha$ -cyclodextrin ( $\alpha$ -CD) and carboxymethyl chitosan (CMS) as dual raw materials. The synthesis involves dispersing these polymers in water, followed by the addition of functional monomers such as acrylic acid (AA) and sodium styrene sulfonate (SS). Under a nitrogen atmosphere and heating, the reaction is mediated by the potassium persulfate (KPS) initiator and N-methylenebisacrylamide (MBA) crosslinker, resulting in a cross-linked structure that combines hydrophobic cavities for pollutant inclusion and ionic networks for metal chelation (Zhang et al., 2026). Each synthesis and cross-linking method presents distinct advantages, disadvantages, and limitations, making the choice of technique specific to each intended application.

### Adsorption properties and mechanisms of waste-derived hydrogels

Hydrogels derived from biomass and industrial residues possess a three-dimensional polymer network structure that endows them with exceptional swelling capacity and high-water affinity (Hossain et al., 2023; Zhou et al., 2025). This porous architecture expands upon contact with the effluents, allowing organic pollutant molecules to rapidly diffuse into the interior of the matrix, where they are encapsulated and retained (Ahmadian et al., 2023; Hossain et al., 2023). Furthermore, synthesis from materials such as paper waste, sugarcane bagasse, or textile wool results in materials with a highly specific surface area and

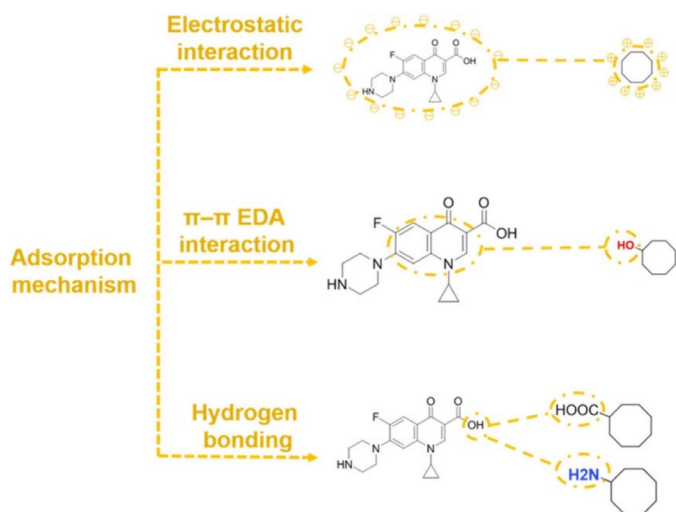
tunable porosity,- fundamental properties for maximizing the active sites available for contaminant capture (Rezvani-Ghalhari et al., 2024; Barzegarzadeh and Asadi, 2025).

The effectiveness of these hydrogels in the removal of organic pollutants, such as antibiotics and dyes, is attributed to the abundance of reactive functional groups on their surface, including hydroxyl (-OH), carboxyl (-COOH), amine (-NH<sub>2</sub>), and sulfonic groups (Mittal et al., 2023; Zhang et al., 2026). In the case of biopolymers such as chitosan and cellulose extracted from waste, these groups enable electrostatic interactions, which represent the dominant mechanism for pharmaceutical removal (Li, D. et al., 2025; Zhou et al., 2025).

The solution pH and the point of zero charge ( $pH_{pzc}$ ) of the material are determinant factors that regulate the electrostatic interactions between the adsorbent and the pollutant (Khumalo et al., 2024; Minaei et al., 2024). When the medium pH differs from the hydrogel's  $pH_{pzc}$ , the material surface acquires a net charge that can attract or repel the contaminant depending on its ionic speciation (Hernandez Monroy et al., 2025; Zhou et al., 2025). For pollutants such as ciprofloxacin, adsorption is optimized in pH ranges where electrostatic attraction forces outweigh those of repulsion, facilitating the binding of molecules within the polymeric network (Gao et al., 2025; Hernandez Monroy et al., 2025).

In addition to electrostatic forces, hydrogen bonding and  $\pi$ - $\pi$  interactions play a crucial role in the adsorption of aromatic and pharmaceutical compounds (Zhou et al., 2025; Zhang et al., 2026). Functional groups such as carbonyl oxygen and amines within the hydrogel act as electron donors or acceptors, forming hydrogen bonds with the hydroxyl or amino groups of antibiotics like sulfamethoxazole (Zhou et al., 2022; Li, D. et al., 2025). Simultaneously, the  $\pi$ -conjugated electron system in carbonaceous residues (such as incorporated biochar) or in the aromatic rings of chemical modifiers allows for stacking interactions with the benzene rings of pollutants, strengthening the chemical binding at the solid-liquid interface (Minaei et al., 2024; Zhang et al., 2026). Compounds like sulfamethoxazole (SMX) and ciprofloxacin (CIP) act predominantly as electron acceptors, while the functional groups present in the hydrogels (amines and hydroxyls) function as donors, favoring the formation of stable complexes at the adsorbent-adsorbate interface (Figure 1; Sun et al., 2020; Prasannamedha et al., 2022).

Physical mechanisms such as pore filling and hydrophobic interactions are also fundamental, especially for the removal of neutral or low-solubility organic pollutants (Li, D. et al., 2025; Su et al., 2025). The rough morphology and the presence of micro and mesopores enable the mechanical capture of molecules whose size is compatible with the pore diameter, an effect known as the “pore confinement effect” (Khumalo et al., 2024; Li, D. et al., 2025). Hydrophobic sections within the polymer chains create domains of low water affinity that preferentially attract the non-polar moieties of drugs such as diclofenac and ibuprofen, effectively minimizing the free energy during adsorption in the system (Alali et al., 2026; Wang, J. et al., 2025).



**Figure 1 – Adsorption mechanisms of ciprofloxacin (CIP) using -NH<sub>2</sub>-functionalized SA/GO hydrogel.**

Source: Sun et al. (2020).

The synergistic use of agro-industrial waste contributes not only to a reduction in production costs but also to the attainment of highly porous matrices rich in cellulose or biochar-derived structures. When these matrices are modified with metallic components (e.g., MgO or Fe), a significant increase in the density of active functional groups is observed, enhancing drug capture and enabling the rapid recovery of the adsorbent via external magnetic fields, eliminating the need for costly filtration. Furthermore, it also facilitates reusability and mechanical stability - properties that impart economic viability of waste-derived hydrogels in scale-up applications.

Studies demonstrate that these materials can retain over 80% of their initial adsorption capacity after multiple chemical regeneration cycles (utilizing solvents such as ethanol or acidic solutions), consolidating them as sustainable solutions within the circular economy framework for wastewater treatment (Huang et al., 2024; Alali et al., 2026). Table 1 summarizes research employing waste-derived adsorbents for pharmaceutical removal, presenting the target contaminants, adsorption capacities (q<sub>e</sub>), and the predominant adsorption mechanisms.

In summary, the combined involvement of electrostatic interactions, hydrogen bonding,  $\pi$ - $\pi$  interactions, hydrophobic forces, and pore-filling mechanisms accounts for the high adsorption efficiencies reported across diverse classes of contaminants. This further underscores the significant potential for tailoring these materials based on the specific target contaminant and the prevailing conditions of the aqueous medium.

### Transition and scale-up for wastewater treatment systems

Wastewater treatment experiments conducted at the laboratory scale frequently yield promising results due to the rigorous control

imposed during the research process to maintain standardized experimental conditions, such as temperature, pH, and contaminant concentration (Ak and Gunduz, 2013). The variability and complexity of industrial or domestic wastewater matrices, fluctuations in organic loading, and the presence of inhibitors can significantly affect treatment efficiency (Saleem et al., 2022). While bench-scale studies operate under controlled conditions and reduced volumes, industrial-scale systems face challenges such as maintaining steady-state operational conditions and managing large volumes of wastewater (Koyuncu and Ariman, 2020).

While laboratory studies are essential for the development and understanding of new wastewater treatment technologies, conducting real-scale tests is fundamental to evaluate the practical viability and performance of these processes under varied operational conditions (Saleem et al., 2022). This integrated approach ensures that the developed technologies are effective and applicable in real-world scenarios, contributing to the continuous improvement of wastewater treatment systems (Ho et al., 2019). Under these circumstances, high pollutant removal rates can be achieved, providing a detailed understanding of the underlying mechanisms involved. Therefore, the scalability of wastewater treatment using hydrogels tested in the laboratory for large-scale application depends on various operational parameters, such as pH, temperature, contact times, and contaminant concentration (Ahmed et al., 2024; X. Tang et al., 2025).

For the transition from laboratory-scale to industrial-scale applications in pharmaceutical wastewater treatment, materials must overcome the mechanical fragility and recovery challenges associated with powdered catalysts. For instance, transforming catalysts and adsorbents into chitosan or alginate-based hydrogel beads promotes uniform dispersion, mitigates aggregation, and enhances contact with organic contaminants in practical scenarios (Ahmaruzzaman et al., 2023; Ruan et al., 2024). These systems offer superior structural stability and mechanical integrity, enabling adsorbents to be recovered and reused without significant material loss via magnetic separation or sieving. These factors are essential for large-scale economic viability (Zhou et al., 2022; Ruan et al., 2024).

The implementation of fixed-bed column systems represents one of the most promising applications for the continuous treatment of large volumes of effluents containing pharmaceuticals and antibiotic resistance genes (ARGs). Scale-up studies have demonstrated that columns dimensioned up to ten times the test volume maintain satisfactory performance, treating substantial quantities of actual wastewater (Pant et al., 2024). Mathematical modeling serves as a crucial tool to predict the breakthrough point and saturation capacity in the beds of MXene-alginate or biochar-MgO-chitosan beads, allowing for the precise design of industrial columns capable of removing ciprofloxacin and naproxen with high efficiency (Prasannamedha et al., 2022; Ghani et al., 2023; Li, Z. et al., 2025).

**Table 1 – Application of hydrogels derived from waste and repurposed materials for the adsorption of pharmaceuticals: experimental conditions, maximum adsorption capacity ( $q_{\max}$ ), and underlying mechanisms.**

| Hydrogel                                               | Target Contaminants                  | Experimental Conditions                        | $Q_{\max}$ (mg/g) | Adsorption Mechanisms                                                                       | Ref.                            |
|--------------------------------------------------------|--------------------------------------|------------------------------------------------|-------------------|---------------------------------------------------------------------------------------------|---------------------------------|
| BC-MgO-CS (Wheat Straw)                                | Ciprofloxacin (CIP)                  | pH 4; 25°C; Dosage: 0.1 g/L; Time: 24 h.       | 1,678.9           | Electrostatic interactions, $\pi$ - $\pi$ , hydrogen bonding, and complexation.             | (Li, D. et al., 2025)           |
| PVA/CMA (Double-Network)                               | Diclofenac sodium (DCFS)             | pH 7; 25°C; Dosage: 0.5 g/L; Time: 72 h.       | 1,012             | Predominantly governed by ion exchange between DCFS anions and chloride.                    | (Wang, L. et al., 2025)         |
| ADS-Gel-1 ( $\alpha$ -CD/CMS) (Carboxymethyl chitosan) | Norfloxacin (NOR)                    | pH 7; 25°C; Dosage: 0.4 g/L; Time: 150 min.    | 861.48            | Host-guest inclusion ( $\alpha$ -CD cavity), $\pi$ - $\pi$ , and H-bonding.                 | (Zhang et al., 2026)            |
| i.Carr@Bent@PTC (Bentonite)                            | Losartan Potassium (LP)              | pH 4.25; 25°C; Dosage: 0.5 g/L; Time: 500 min. | 467.61            | Complexation, $\pi$ - $\pi$ , and electrostatic interaction.                                | (Ijaz et al., 2024)             |
| GO/MIL-88/Alg (Alginate)                               | Doxycycline (DOX)                    | pH 4; 25°C; Dosage: 5 g/L; Time: 1,440 min.    | 131.57            | Synergistic effect between MOF/Graphene; electrostatic interactions and $\pi$ - $\pi$ .     | (Karimi and Namazi, 2024)       |
| SACFe (Sugarcane Bagasse)                              | sulfamethoxazole (SMX)               | pH 6.2; 25°C; Dosage: 0.6 g/L.                 | 58.44             | Heterogeneous; chemisorption, EDA interactions, and H-bonding.                              | (Prasannamedha et al., 2022)    |
| PBC (Sludge-based Biochar)                             | sulfamethoxazole (SMX)               | pH 4; 25°C; Dosage: 2 g/L; Time: 24 h.         | 45.6              | $\pi$ - $\pi$ , EDA interactions and charge-assisted hydrogen bonding.                      | (Minaei et al., 2024)           |
| PBC (Sludge-based Biochar)                             | Lincomycin (LIN)                     | pH 3; 25°C; Dosage: 2g/L; Time: 72h.           | 26.6              | Electrostatic Interactions, hydrogen Bonding and Van der Waals and Hydrophobic Interactions | (Minaei et al., 2024)           |
| Cellulose/MMO/PVA (newspapers)                         | Ciprofloxacin (CIP)                  | pH 4.5; 25°C; Dosage: 0.55 g/L; Time: 83 min.  | 19                | Multilayer adsorption (Freundlich) and chemisorption.                                       | (Rezvani-Ghalhari et al., 2024) |
| MX@SA (MXene/Alginate)                                 | Naproxen (NPX)                       | pH natural; 25 °C; Leito fixo (coluna).        | 22                | Multisite, $\pi$ - $\pi$ , EDA interactions and inner-sphere coordination.                  | (Ghani et al., 2023)            |
| NH <sub>2</sub> -DN (Graphene/Alginate)                | Ciprofloxacin (CIP)                  | pH 7; 25°C; 150 rpm; Time: 120 h.              | 301.36            | Chemical affinity, hydrogen bonding, $\pi$ - $\pi$ and EDA interactions                     | (Sun et al., 2020)              |
| HBP-DAC@SA (Diacetal cellulose)                        | Levofloxacin (LEV)                   | pH 5; 30°C; Dosage: 0.2 g/L; Time: 90 min.     | 216.72            | Monolayer chemisorption; 3D adsorption network with high site density.                      | (Tao et al., 2025)              |
| TG-C12 Br (Tragacanth Gum)                             | Diclofenac sodium (DCF)              | pH 5; 30°C; Dosage: 0.6 g/L.                   | 327.87            | Electrostatic attraction, hydrophobic interactions, H-bonding, and $\pi$ - $\pi$ .          | (Du et al., 2024)               |
| Fe(III)-Alginate (Alginate)                            | Doxorubicin Hydrochloride            | pH natural; 37°C; Time: 24h                    | 3.897             | Hydrogen bonding, electrostatic attraction, and coordination.                               | (Bahwal et al., 2025)           |
| MSAC@GE-SA (Maize Stalks)                              | Doxorubicin Hydrochloride (Doxo.HCL) | pH natural; 25°C; Dosage: 1.2 g/L; Time: 24 h. | 239.41            | Electrostatic interactions, pore filling, H-bonding, $\pi$ - $\pi$ , and EDA.               | (Devre and Gore, 2023)          |

The integration of advanced oxidation processes (AOPs) with large-scale hydrogel beds enhances the in-situ degradation of persistent pharmaceuticals, overcoming the limitation of merely transferring pollution from the liquid to the solid phase. Hybrid systems employing sonocatalysis or heterogeneous Fenton processes in the presence of Fe<sub>3</sub>O<sub>4</sub>/AC/CH beads achieve significant reductions in COD and toxicity within the first hour, turning toxic contaminants into less harmful byproducts (Zhou et al., 2022; Verma and Moholkar, 2025). Furthermore, beads functionalized with graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) enable bed regeneration via ultraviolet or solar light,

reactivating adsorption sites and degrading adsorbed pollutants, which simplifies operations in industrial facilities (Zhou et al., 2022; Hernandez Monroy et al., 2025).

Another frontier in scale-up applications is the use of photobioreactors and “living hydrogels” for sustainable bioremediation. Thin-layer cascade reactors operated outdoors have demonstrated the technical feasibility of removing pharmaceuticals such as sulfamethoxazole, ofloxacin, and diclofenac from real wastewater, while biomass productivity remains unaffected even under chemical stress (Morillas-España et al., 2025). Simultaneously, the development of nanocellulose-based

hydrogels embedded with fungi such as *Rhizopus oryzae* offers a mechanical self-reinforcement approach, where the microorganism utilizes nutrients from the effluent to proliferate and strengthen the gel network, resulting in significant reductions of BOD and COD in complex industrial wastewater (X. Tang et al., 2025).

The commercial viability of these systems depends on techno-economic assessments that demonstrate efficiency in comparison to traditional methods. Recent research indicates that biochar-iron oxide catalysts encapsulated in chitosan-based hydrogel beads can process hundreds of bed volumes of real-world wastewater with competitive costs of approximately \$3.42 per treated ton (Ruan et al., 2024). The use of biomass residues, such as sugarcane bagasse for the synthesis of magnetic sodium alginate beads (SACFe), is able to reduce the manufacturing costs and simplify operations, promoting a circular economy where agro-industrial byproducts remove pharmaceutical pollution in an economical and sustainable manner (Prasannamedha et al., 2022; Ruan et al., 2024).

Finally, the transition of hydrogel-based wastewater treatment from laboratory scale to industrial applications requires overcoming operational, structural, and economic challenges associated with the complexity of real-world matrices and the need for long-term stability. Recent evidence demonstrates that strategies such as adsorbent granulation into hydrogel beads (Ahmaruzzaman et al., 2023; Ruan et al., 2024), the implementation of fixed-bed column systems (Pant et al., 2024), the integration with advanced oxidation processes (Zhou et al., 2022; Verma and Moholkar, 2025), and innovative biotechnological approaches (Morillas-España et al., 2025; X. Tang et al., 2025) render this scalability technically viable and economically competitive. Thus, when combined with robust techno-economic assessments and the use of waste-derived precursors as raw materials, these technologies can be consolidated as promising and sustainable alternatives for the large-scale advanced treatment of pharmaceutical wastewater.

## Bibliometric analysis

The search conducted using the specified keywords (“Hydrogel”, “Pharmaceuticals wastewater treatment”, “Adsorption of pharmaceuticals”, “Waste-derived hydrogel”, and “Sustainable hydrogels”) retrieved 703 articles from the Web of Science database, while the same search yielded 71 articles in the Scopus database. Across both databases, all retrieved articles underwent a preliminary screening to ensure that only those demonstrating scientific relevance and quality are included, resulting in a final sample of 315 articles from the Web of Science platform and 65 from Scopus.

The selected records were exported in BibTeX format and subjected to integrated analysis in the R software using the Bibliometrix package. This step aimed to identify and remove duplicate entries while consolidating information retrieved from the different databases. Following this procedure, a total of 373 documents were included in the final analysis. Figure 2 presents the schematic representation of the methodology applied for bibliometric analysis.

Figure 3 presents the temporal evolution of publications from 2017 to 2026, highlighting the primary research trends related to the use of hydrogels in wastewater treatment.

Bibliometric analyses identified approximately 96 studies employing agro-industrial residues as raw materials or fillers in the synthesis of adsorbent hydrogels, highlighting the vast diversity of biomasses investigated. Abundant plant residues predominate, such as soybean residue (Wu et al., 2022), sugarcane bagasse (Prasannamedha et al., 2022), rice husk and straw (Nath et al., 2025), corn stalk (Devre and Gore, 2023), and lignocellulosic waste from the paper and pulp industry (Mondal et al., 2021; Luong et al., 2024; Radoor et al., 2024). These residues reflect the potential for producing functional hydrogels aligned with the circular economy principles.

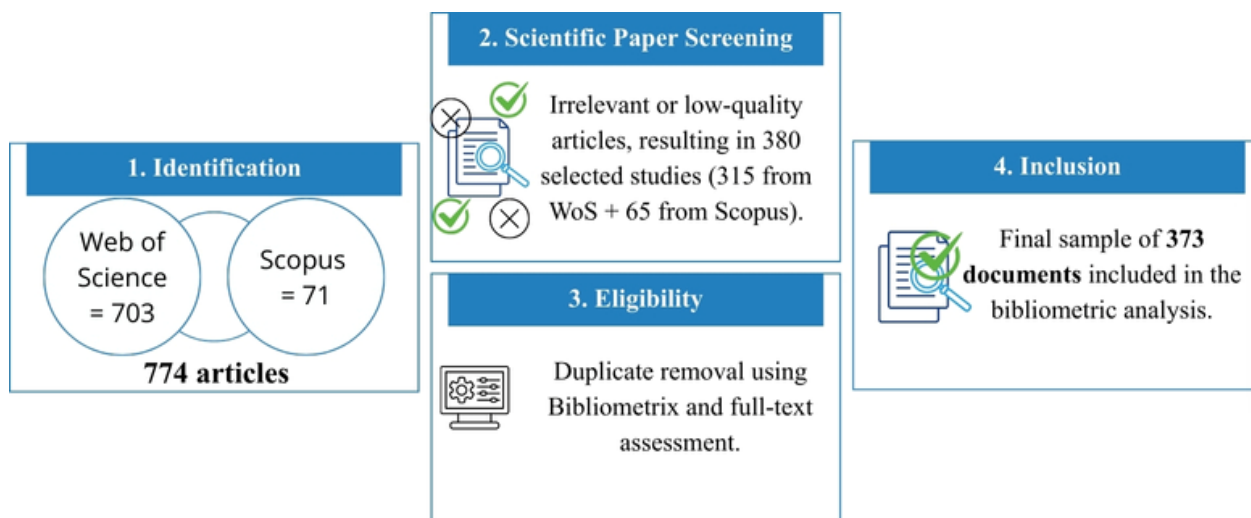
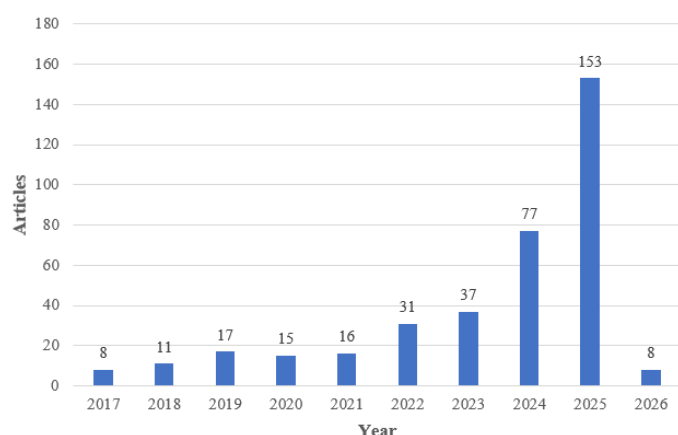


Figure 2 – Schematic representation of the methodology applied for bibliometric analysis.

Despite the diversity of reported biomasses, only approximately 32 studies explicitly address the removal of pharmaceutical contaminants, indicating that this application remains a minority compared to the traditional uses of these materials. The most extensively investigated pharmaceuticals include sulfamethoxazole (Prasannamedha et al., 2022), ciprofloxacin (Li, D. et al., 2025), tetracycline (Huang et al., 2024), levofloxacin (Tao et al., 2025), amoxicillin (Roa et al., 2025), doxycycline (Huang et al., 2024), diclofenac (Du et al., 2024), paracetamol (Elessawy et al., 2024), ibuprofen (Chan et al., 2021), naproxen (Ghani et al., 2023), and doxorubicin (Devre and Gore, 2023). These results indicate that, although agro-industrial residues exhibit high adsorbent potential, their targeted application for pharmaceutical removal still represents a significant gap in the literature, reinforcing the relevance of this review's scope.

In contrast to waste-based systems, approximately 90 articles utilize hydrogels synthesized from commercial polymers or purified biopolymers, such as polyacrylamide (PAM), polyacrylic acid (PAA), and polyvinyl alcohol (PVA), among others (Wang, L. et al., 2018; Gokhale et al., 2022; Mota et al., 2023; İlyasoğlu et al., 2025; Mosaffa et al., 2026). These materials dominate environmental applications due to their high reproducibility, mechanical stability, and ease of functionalization. However, this predominance highlights a significant reliance on conventional matrices at the expense of waste-derived approaches, particularly regarding the removal of pharmaceuticals.



**Figure 3 – Articles published on the Web of Science and Scopus databases using the keywords: “Hydrogel”, “Pharmaceuticals wastewater treatment”, “Adsorption of pharmaceuticals”, “Waste-derived hydrogel”, and “Sustainable hydrogels”.**

**Table 2 – Summary table of the integrated bibliometric analysis.**

| Analyzed Axis                                      | Main Focus                                       | Articles* |
|----------------------------------------------------|--------------------------------------------------|-----------|
| Waste-derived hydrogels                            | Diverse agro-industrial residues                 | 96        |
| Waste-derived hydrogels for pharmaceutical removal | Antibiotics and anti-inflammatories              | 32        |
| Conventional polymer hydrogels                     | Synthetic polymers and commercial biopolymers    | 90        |
| Hydrogels for the removal of other contaminants    | Metals, dyes, nutrients, oils, and microplastics | 190       |

\*The values are not mutually exclusive and represent approximations, as an individual study may encompass multiple categories of materials or investigate several different contaminants.

More broadly, the literature reveals that the use of hydrogels in the treatment of non-pharmaceutical contaminants accounts for the majority of applications, totaling approximately 190 articles. Most of these studies focus on the removal of heavy metals, industrial dyes, and nutrients (Tummino et al., 2020; Kaur et al., 2025; Mahata et al., 2025; Li et al., 2026). Other applications include oil–water separation and the removal of hydrocarbons, pesticides, and microplastics (Cao et al., 2025; Patel et al., 2025; Zhang et al., 2025; Sharma et al., 2026). Comparatively, the data confirm that pharmaceutical removal still occupies a secondary position, highlighting the need to expand investigations that integrate agro-industrial residues and pharmaceutical contaminants, especially in light of the environmental risks associated with these compounds. Table 2 presents a compilation of the bibliometric analysis.

This bibliometric analysis reinforces that, although hydrogels are extensively explored for environmental contaminant remediation, the bulk of scientific production remains concentrated on applications involving conventional polymers and non-pharmaceutical contaminants, such as heavy metals and dyes. In contrast, the use of agro-industrial waste-derived hydrogels for pharmaceutical removal still represents a limited fraction of the literature, despite the high potential of these materials in terms of sustainability, availability, and functional diversity. These findings confirm that the integration of waste valorization and the removal of pharmaceutical contaminants constitute a significant research gap, highlighting the need for studies that advance the understanding of adsorption mechanisms, performance evaluation in real matrices, and large-scale application viability.

## Conclusions

This study addresses the possibilities of utilizing agro-industrial waste-derived hydrogels for the treatment of pharmaceutical effluents. Through the analysis of physicochemical properties, adsorption mechanisms, synthesis and modification methods, and treatment performance, it is evident that hydrogels exhibit a high potential for the removal of pharmaceutical contaminants, offering benefits in terms of efficiency, sustainability, and cost-effectiveness.

The increasing use of agro-industrial residues in the development of novel materials facilitates the reduction of production costs and promotes the circular economy, providing a sustainable and value-added solution. Furthermore, to ensure efficiency, it is evident that the chemical and physical modifications of these hydrogels, such as the introduction of functional groups and thermal treatments, significantly enhance their adsorption capacity, enhancing their effectiveness for the treatment of diverse pharmaceuticals.

On the other hand, for the large-scale implementation of hydrogels to be achieved, several research gaps must be addressed. The regeneration of hydrogels following pharmaceutical adsorption and the management of their final disposal remain critical issues that require further investigation to ensure the long-term viability of this technology. Another identified gap is the transition from laboratory-scale results to applications in real treatment systems, where factors such as pH, temperature, and the presence of competing contaminants in actual effluents must be evaluated collectively. Furthermore, the integration

of hydrogels with other treatment technologies represents a promising strategy to enhance pharmaceutical removal efficiency.

In this context, the exploration of novel waste precursors, refinements in hydrogel modification and regeneration methodologies, the assessment of their life-cycle cost as well as environmental impact will enhance hydrogel efficiency. This constitutes a promising solution to the challenge of pharmaceutical pollution and represents a significant advancement toward sustainability in waste management and effluent treatment.

### Authors' Contributions

**Brandler**, D.: conceptualization; data curation; methodology; formal analysis; investigation; software; writing – original draft. **Pinto**, Y. C.: investigation; methodology; writing – review & editing. **Conceição**, B. M.: investigation; methodology; writing – review & editing. **Korf**, E. P.: conceptualization; supervision; project administration; data curation; resources; writing – review & editing. **Pasquali**, G. D. L.: conceptualization; supervision; project administration; data curation; resources; writing – review & editing.

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