Adsorption-based removal of amoxicillin from aqueous environments: a mini review

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Abstract. Amoxicillin's prevalence in aquatic environments, stemming from widespread medical usage, serves as a significant indicator of pharmaceutical contamination. Adsorption stands out as the preferred method for addressing this issue due to its simplicity, efficacy, practicality, and cost-effectiveness. This systematic review delves into peer-reviewed literature on amoxicillin removal through adsorption, drawing from databases like ScienceDirect and Scopus. Researchers have investigated adsorption equilibrium under varied conditions, exploring parameters such as pH, temperature, and adsorbent dosage. The diverse range of observed elimination levels underscores the critical importance of careful adsorbent selection, with capacities spanning from 10 to 1500 mg/g. Pseudo-second-order kinetic models and the Langmuir isotherm model frequently offer suitable descriptions of experimental data. Future research avenues could explore alternative kinetic models to deepen our understanding of amoxicillin adsorption mechanisms and foster the development of innovative adsorbents. ***Keywords :*** Adsorption, Amoxicillin, Kinetic model, Isotherm.

1 Introduction

Pharmaceuticals, personal care products, and other chemical substances are categorized as emerging pollutants, sparking growing interest within the scientific community in recent years [1–3]. These emerging pollutants are defined by the United States Environmental Protection Agency (USEPA) as new chemical compounds lacking regulatory status concerning their environmental and human health impacts [4]. The use of medications and personal care products has significantly increased in recent times. Among these medications, amoxicillin ($C_{16}H_{19}N_3O_5S$, CAS 26787-78-0) (Fig.1), is widely used as an antibiotic in both human and veterinary treatments, accounting for between 50 and 60% of the total global antibiotic production [5]. Studies have shown that over 80% of amoxicillin is excreted in human urine within the first two hours after ingestion, with a portion of its active form not completely metabolized. This results in its presence in effluents from wastewater treatment plants at concentrations ranging from 170 to 1270 ng/L [6–8], in surface waters between 120 and 200 ng/L [9], and in hospital effluents, reaching up to 900 ng/L in Australia [10].

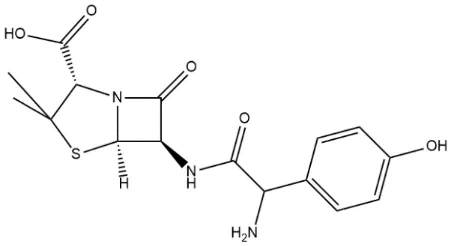

Fig. 1. Chemical structure of Amoxicillin (AMX).

To remove amoxicillin from wastewater, several methods have been devised. Among these, adsorption stands out for its significant advantages, including its efficiency, reduced operational cost, high selectivity, and ease of handling [11].

2 Amoxicillin removal through adsorption

Adsorption is a process of mass transfer wherein a solute migrates from the fluid phase to the surface of a solid, termed an adsorbent [12,13]. This adherence can arise from physical forces, known as physisorption, involving interaction through Van der Waals forces, or from chemical bonding, termed chemisorption, wherein molecules and atoms bond to the adsorbent surface via chemical bonds,

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typically of a covalent type. In chemisorption, they tend to bind to sites offering the highest coordination with the solid [14–16]. In aqueous media, adsorption is a competitive process whose equilibrium is determined by numerous factors arising from the properties of the adsorbent, the adsorbate, and the solvent [15,17]. The main factors influencing adsorption equilibrium include the porous structure of the solid, energetic heterogeneity, and the chemical properties of the surface, such as the presence of functional groups [18]. Additionally, the adsorption phenomenon depends on the chemical properties, structure, and mutual interactions, as well as interactions with the solid surface, of both the adsorbate and the solvent [17,19]. Other variables, such as contact duration, concentration of solute, temperature and pH, can also affect the adsorption capacity [20]. Consequently, comprehensive studies involving kinetics, isotherms, and thermodynamics are imperative for elucidating adsorption equilibrium [20].

2.1 Adsorbents evolution

The porous material utilized in the adsorption process may vary in nature, being either organic, like activated carbon, biochar, and biosorbents, or inorganic, as seen in aluminosilicates, or a combination of both, as observed in composite materials [21]. Literature research has revealed the use of various materials for the removal of amoxicillin by adsorption. Among these materials are activated carbons [22–25], including modified versions [26], as well as clay [27,28], carbon nanotubes [23,29,30], nanocomposites [31–33], graphene oxide [34], magnetic graphene oxide, and various forms of biomass such as palm bark [35], wheat grains and their modified variants [36], chitosan beads [37], as well as almond shell ashes [38]. Through bibliographic analysis, the evolution of adsorbents over the past years has been traced. Alternatives have emerged to replace powdered activated carbon and granular activated carbon in the adsorption of amoxicillin. Researchers have explored materials such as clays, biosorbents, biochar, and more recently, composites and nanomaterials. This shift can be attributed to the growing demand for activated carbon, resulting in an increase in its price. Another factor limiting the widespread use of activated carbon is the challenge in its regeneration, with estimates suggesting that less than 40% of activated carbon impregnated with organic compounds can be recovered [39].

Furthermore, it is essential to develop more porous materials with an increased surface area. The appropriate choice of adsorbent typically constitutes the crucial first step in the adsorption process [40]. The solute adsorption capacity is closely linked to the surface area of the adsorbent, where a larger surface area and pore volume can result in higher adsorption capacity [15].

2.2 Influence of adsorbent dosage, pH and temperature

The adsorption process is influenced by various factors, including the pH, temperature of the medium, the presence of ions, the initial concentration of the pollutant, and the amount of adsorbent in contact with the pollutant at any given time [41]. According to the literature review, it can be inferred that the pH of the solution, temperature, and the amount of adsorbent used are likely the key factors affecting the absorption of AMX onto the adsorbent [42]. The objective of this section is to provide a detailed description, as indicated in Table 1, of these parameters involved in AMX adsorption studies. Each of them is examined and discussed individually throughout the text.

2.2.1 Effect of initial pH

The pH level of the solution impacts the adsorption process of amoxicillin, involving various mechanisms such as cation exchange and π-π interactions [34,43,44]. Amoxicillin's charge fluctuates with pH changes, influencing its adsorption capacity on different adsorbents. At pH levels between 3 and 6, amoxicillin adopts a zwitterionic form, facilitating its adsorption. However, at higher pH levels, the dual negative charge of amoxicillin hinders its adsorption due to repulsive forces [45]. Apart from electrostatic interactions, other mechanisms like π-π interactions, notably observed with magnetic graphene oxide [34], may also contribute. Additionally, certain adsorbents, such as activated carbon derived from guava seeds, demonstrate exceptional adsorption performance across various pH ranges (Fig. 2), indicating the involvement of diverse mechanisms like hydrogen bonding and donor-acceptor electron interactions [45].

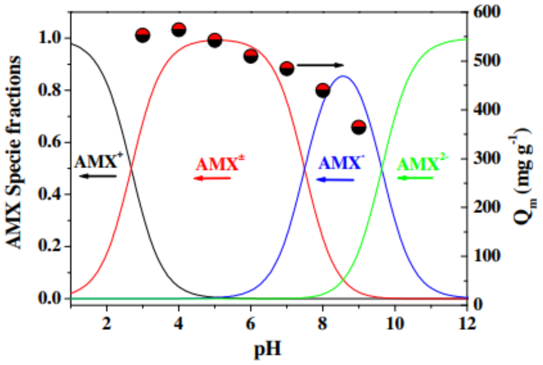

Fig. 2. The pH-dependent distribution of AMX species and its influence on the adsorption of AMX onto activated carbon derived from guava seeds. [45].

2.2.2 Effect of temperature

The influence of temperature on the adsorption process is a crucial factor to consider, affecting the capacity of adsorbents to remove amoxicillin. Temperature increase can lead to a reduction in the effectiveness of certain adsorbents, as observed with modified wheat grains (Fig. 3), bentonite, and activated carbon [22,36,46]. This trend suggests an exothermic adsorption process, as indicated by the studies of Boukhelkhal (2016) [36], Budyanto (2018) [22], and Chitongo (2019) [46]. The decline in effectiveness at elevated temperatures could be due to reduced interactions between amoxicillin and the active sites present on the adsorbent surface. [36,46].

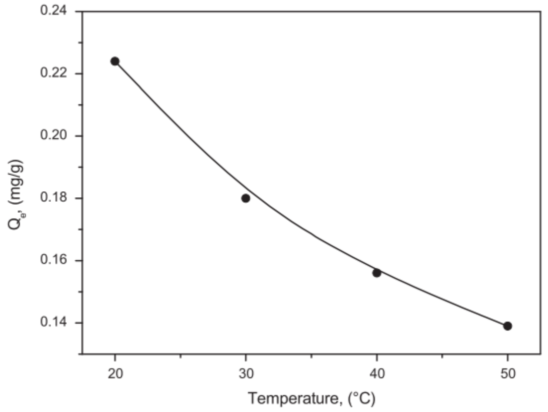

Fig. 3. Effect of the temperature on Amoxicillin adsorption onto modified wheat grains [36].

On the other hand, temperature increase can enhance adsorption efficiency for some adsorbents, such as graphene oxide and magnetic graphene oxide, as well as bentonite modified with hexadecyltrimethylammonium chloride [34,44]. This observation, supported by the studies of Moradi (2015) [34] and Zha (2013) [44], suggests that adsorption in these cases is primarily driven by entropy.

2.2.3 Effect of dosage of adsorbent

The addition of adsorbent can significantly influence its adsorption capacity by altering the available surface area and the number of binding sites. Increasing the adsorbent dosage generally tends to improve amoxicillin removal, although this response may vary depending on the specific material used [23,29,38,44,46,47,48]. Results show that as the dosage increases, certain materials exhibit improved adsorption performance. Optimal doses for these materials are observed at 0.1 g/L for magnetic graphene oxide and 4 g/L for wheat grains, respectively, while others, such as almond shell ash and activated carbon (Fig. 4), show a decrease in efficiency at higher dosages [24,34,36]. These differences can be attributed to particle aggregation phenomena [49,50], reducing the surface area available for adsorption [24]. In summary, optimizing adsorbent dosing is crucial to maximize its effectiveness in removing amoxicillin.

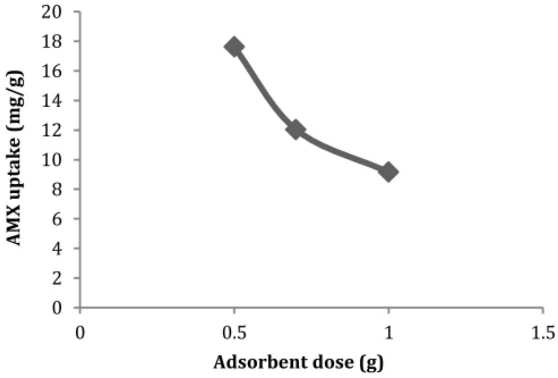

Fig. 4. Effect of adsorbent dosage on AMX uptake onto activated carbon [24].

2.3 Equilibrium, kinetic modeling and thermodynamic studies

The utilization of adsorption isotherms serves to demonstrate the connection between the adsorbed pollutant on the adsorbent and the pollutant concentration within the solution [51]. Investigating isothermal models constitutes a crucial aspect of adsorption research, elucidating the interactions between contaminants and adsorbents [51]. The most widely used isotherms for modeling amoxicillin adsorption are Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich. The Langmuir isotherm model fits well the experimental data. The Freundlich isotherm model is also used for modeling amoxicillin adsorption. The Sips model is found to be better fitted compared to Langmuir and Freundlich models. These findings are based on studies by Balarak (2017) [23], and De Franco (2017) [25], as well as the comprehensive review by Majd (2022) [52].

Studying kinetics is crucial for predicting optimal conditions in full-scale adsorption processes [53], offering insights into adsorption mechanisms and potential rate-controlling steps [53]. Among the commonly utilized models for AMX adsorption are the Pseudo-first order, Pseudo-second order, Elovich, and Avrami.

Notably, the Pseudo-second-order model effectively interprets experimental kinetic data. Thermodynamic analyses are employed to determine parameters like Gibbs free energy (ΔG°), enthalpy change (ΔH°), and entropy change (ΔS°), which also aid in understanding the adsorption mechanism [51].

3 Conclusion

In summary, the study of amoxicillin adsorption reveals significant considerations for the remediation of water contaminated by this pharmaceutical compound, particularly due to its prevalence as an emerging pollutant capable of causing serious environmental and human health issues. The diversity of adsorbents employed, such as biomass, carbon nanotubes, nanocomposites, clay, and metal-organic frameworks, underscores the importance of this research in evaluating their efficacy in removing amoxicillin from wastewater. It is critical to note that while chemical modification of adsorbents may enhance their adsorption capacity, it also poses risks of secondary pollution and increased treatment costs, emphasizing the need for thorough evaluation of the pros and cons of such modifications. Kinetic and thermodynamic studies provide crucial insights into adsorption mechanisms and optimal treatment conditions. However, understanding of interaction mechanisms remains limited, necessitating further research to assess the application and effectiveness of the studied adsorbents in real-world systems, such as wastewater and treatment plant

Table 1 : A synopsis of several investigations concerning the adsorption of AMX onto various adsorbents.

Adsorbent	**Agent modifying**	**Initial concentration (mg/L)**	**Adsorbent dosage (g/L)**	**Adsorption capacity (mg/g)**	**Adsorption kinetics**	**Isotherms**	**Time (min)**	**pH**	**T (°C)**	**Ref**
Actived carbon from guava	NaOH	50 – 800	1.0	570.48	Elovich	Redlich-Peterson	240	4.0	25	[45]
Bentonite	–	300	0.1 - 1.5	–	Pseudo-second order	Langmuir/ Freundlich	480	2.31	30	[43]
Wheat grains	Tartaric acid	240	4	31.25	Pseudo-second order	Temkin	5	7.0	25	[36]
Magnetic graphene oxide	–	10 – 100	0.1	372.4	Pseudo-second order	Langmuir	180	6.0	30	[34]
Graphene oxide	–	10 – 100	0.1	280.8	Pseudo-second order	Langmuir	180	6.0	30	[34]
Actived Carbon from coffe waste	–	50 – 450	1.0	370.0	Pseudo-second order	Sips/ Langmuir	–	10.0	–	[54]
Ferrihydrite	–	20 – 200	0.66	75.0	–	Liu	–	4.0	25	[55]
Actived carbon from cistus shell	H_3PO_4	100 – 800	–	1250.0	Pseudo-second order	Langmuir/ Freundlich	–	–	–	[56]
Montmorillonite	Dodecyl dimethyl benzyl ammonium chloride	10 – 180	0.1	13.29	Pseudo-second order	Freundlich	240	9.0	25	[57]
Actived carbon carbon from dried pomegranate wood	NH_4Cl	500	0.8	438.6	Pseudo-second order	Langmuir	360	6.0	25	[58]
Palm bark	–	10 – 100	3.0	35.92	–	Langmuir	90	–	25	[35]

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