Mini Review

New Techniques Used for Removing Antibiotic Residues and Antibiotic Resistance Genes from Water

HATASO, USA
New Techniques Used for Removing Antibiotic Residues and Antibiotic Resistance Genes from Water

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Abstract
Overuse and misuse of different antibiotics are considered as one of the main causes of antibiotics accumulation in the environment, most commonly used antibiotics are semimetabolized and excreted by humans to the environment. Studies reported that antibiotic usage exceeds 100,000 tons per year, and this amount may be shocking. However, in fact, the persistence of antibiotic compounds may be more dangerous than the used amount, so it is necessary to develop new methods for elimination of these new pollutants from the environment, especially from water. In this paper, we highlight new and more efficient methods used for removing antibiotic residues (AR) and antibiotic resistance genes (ARGs). The new techniques are Fe₃O₄/red mud nanoparticles, 3D hierarchical porous-structured biochar aerogels, calcined layered double hydroxides, co-doped UiO-66 nanoparticles, Cu@TiO₂ hybrids, bioelectrochemical systems, and aerobic granulation process. Most of these methods showed good performance in removing AR and ARGs that ranged from 85% to 95%. These percentages are consider very efficient compared with traditional wastewater treatment methods.

Keywords: ARGs; Antibiotics removing; Antibiotics adsorption.

1. INTRODUCTION

In the past decades, the term “water pollution” was restricted with chemicals (organics and inorganics) such as fertilizers, toxic heavy metals, industrial wastes, and pesticides, and microbial pollutants such as pathogenic microbial cells. However, recently, with expansion of industrialization and continuous urban development, a new type of chemical pollution appeared because of pharmaceutical products. This new pollution is caused by antibiotic pollutants and pollution of water by antibiotic resistance genes (ARGs) [1-5].

Recent studies report that antibiotic usage exceeds 100,000 tons per year, and there is increasing concern over the fate of these substances because a large number of antibiotics cannot be metabolized completely in human and animals [6]. The topics of antibiotics and ARGs are growing worldwide [7], and they are classified as a new class of water contaminants because of their potential adverse effects on aquatic ecosystems and human public health [8].

Water environment that receives urban and medical effluents is particularly threatened by antibiotic pollutants due to poor treatment and the presence of pharmaceuticals in the final step of wastewater treatment plant (WWTP), which finally discharges into aqueous ecosystems. It may be related with the emergence and spread of multidrug-resistant pathogenic bacteria, and it can lead to unpredictable ecological impacts and responses, and may have an impact on human health [1, 9].

In the past, effluent water of WWTP was discharging directly into the aqueous environment without using developed techniques for removing antibiotic residues (AR) and ARGs, and this behavior is considered as the main source of AR and ARGs spreading in the environment, which finally leads to accumulation and increase of antibiotics resistance bacteria (ARB) and ARGs.

Moreover, studies report that the antibiotics can resist biodegradation due to their antimicrobial nature. For this reason, antibiotics have been classified as emerging pseudo-persistent organic pollutants for their continual input into the environment and permanent presence.

In addition, G. Zhai has reported that AR cannot be efficiently removed by existing traditional wastewater treatment methods applied in WWTPs, which include both biological processes methods (e.g., activated sludge and anaerobic digestion) and physiochemical methods (e.g., coagulation, sedimentation, and filtration) [10].

For these reasons, we will highlight new, modified, and developed techniques that can be used for removing and minimizing AR and ARGs in water before draining wastewater into the aqueous environment.
2. MAIN TECHNIQUES FOR REMOVING AR & ARGs

With increase of AR pollution in wastewater with poor traditional WWT methods, there is an urgent need to use new techniques for removing AR; for this reason, we will explain new and sufficient methods for elimination of antibiotics from water.

2.1. Magnetic Fe₃O₄/Red Mud Nanoparticles

In this method, two combinations of magnetically separable Fe₃O₄-red mud nanoparticles (Fe₃O₄-RM-NPs) are used to remove antibiotics from wastewater. For this method, ciprofloxacin antibiotic was selected to determine the model’s removal efficiency of these two compounds.

Iron is mainly used because of its magnetically separable properties; whereas, red mud is used because of its chemical and mechanical stability, and it acts as a nanomaterial and possesses effective adsorbent properties alternative to commercial adsorbents [11, 12].

Many conditions influence the efficiency of this method; they are red mud amount, pH, contact time, and dosage of Fe₃O₄-RM-NPs. The increase of red mud amount, contact time, and dosage of Fe₃O₄-RM-NPs leads to high removal efficiency. In contrast, the increase in pH leads to decrease in removal efficiency as shown in Figure 1.

According to the results of this method, the removal efficiency was ~90% for all antibiotics. Finally, this method is a successful way to eliminate antibiotics from wastewater.

2.2. 3D Hierarchical Porous-Structured Biochar Aerogel

Hierarchical porous-structured biochar aerogel (3D-PBA) method is designed to remove a specific group of antibiotics called phenicol antibiotics (PAB), which includes thiamphenicol, chloramphenicol, and florfenicol. The main reason for using 3D-PBA is the porous structure of this material showed in Figure 2, which is characterized with three-dimensional imaging giving it unique effects on the water retention properties [13], and because it has high adsorption specificity toward the target PAB [14].

Liu et al. (2019) reported that the 3D-PBA can act as an efficient candidate for ultrafast PAB removal.

2.3. Calcined Layered Double Hydroxide

Calcined layered double hydroxide (CLDH) method designed by [15] is used to remove sulfamethoxazole (SMX), which belongs to the family of sulphonamide drugs. It is commonly used to treat digestive infections and urinary tract infections in humans and animals, so the amount of antibiotics that can be discharged into urban wastewater is considered high. Ciprofloxacin is one of the most commonly prescribed antibiotics and is frequently detected in surface water ecosystems [16].
The layered materials, positive charge on the layer, substitution of divalent by trivalent cations, and anionic characteriza-
tion make CLDH a good material for adsorbing antibiotics from water [17, 18].

Experimentally, the removal efficiency of CLDH for antibiotics depends on the percentage of SMX remaining after a
series of retention cycles. CLDH can remove about 93% of sulfamethoxazole from water depending on the experiment condi-
tions such as pH, temperature, and dosage.

Recently, CLDH is considered a highly preferred material for removing SMX, and it is a promising material for eliminat-
ing widespread pollutants.

2.4. Co-doped UiO-66 Nanoparticle
This technique is proposed by [19]. Co-doped UiO-66 is a novel recyclable nanoparticle used to deactivate tetracycline (TC)
drugs. Co-doped UiO-66 is characterized by high adsorption capacity and photocatalytic performance. The sensitivity of TC to
light Co-doped UiO-66 is used to remove it from water by photodegradation properties simulative with sunlight illumination
(Figure 3); therefore, this method is considered the ideal solution for removing TC.

The investigations by [19] on pharmaceutical wastewater showed high TC removal efficiencies of ~87.1%

2.5. Cu@TiO2 Hybrids
This method is composed of two parts with dual activity: Cu nanoparticles and TiO2 nanoaggregates. Cu nanoparticles have
both adsorption and photocatalytic performance, which are advantageous for removing toxic molecules such as antibiotic
pollutants. TiO2 nanoaggregates also have photocatalytic and absorbency effects [20] because of their porous structure, which
could enhance the adsorption capability [21, 22]. In fact, TiO2 alone shows good effect but if combined with Cu the activity
will increase by five times than that of TiO2 alone. Consequently, the purpose of combining between these two particles is to
produce a hayride with dual action to remove antibiotics. This hybrid prepared by [23] (Figure 4) is used for the degradation
of ciprofloxacin.
Experimentally, Gan et al. (2018) observed that 1.0-Cu@TiO\textsubscript{2} sample could remove nearly 50% of the ciprofloxacin from the solution through adsorption or photocatalysis. Increasing the Cu content, the adsorption capability of the 1.0-Cu@TiO\textsubscript{2}, 5.0-Cu@TiO\textsubscript{2}, 10-Cu@TiO\textsubscript{2}, 20-Cu@TiO\textsubscript{2}, and 50-Cu@TiO\textsubscript{2} samples increased as 173, 229, 258, 241, and 234 mg/g, respectively. This means that the removing efficiency increases with increase in the hayride concentration. The 10-Cu@TiO\textsubscript{2} sample showed the optimal adsorption capability toward 80 mg/L of ciprofloxacin solution.

2.6. Bioelectrochemical Systems
Bioelectrochemical systems (BESs) coupled with microbial metabolisms and electrochemical redox reactions are considered promising alternatives for degradation of ARGs.

This method is composed of microbial fuel cells (MFCs) and microbial electrolysis cells; this technique is ideal for removing ARGs [24].

2.7. Aerobic Granulation Process
This method is active on both AB and ARG. The main mode of action is bioadsorption and biodegradation due to the high protein content and tightly bound extracellular polymeric substances (TB-EPS) of aerobic granular sludge (AGS). The high protein content improves the adsorption capacity among hydrophobic particles [25]. The results revealed that the ARB (main source of ARGs) removal rate increased up to 89.4%. The results confirm that AGS is a good method to remove ARGs [26].

3. CONCLUSION
The primary objective of this review was to highlight ideal methods for removing AR and ARGs from water environment due to the urgent requirement to minimize the problem of antibiotic resistance between microbial cells that are found in water environment. According to previously explained methods, we conclude that most of these methods showed efficient performance to remove AR and ARGs. The removing efficiency of all methods was high, and it ranged from 85% to 95%; this percentage reflects the efficiency of the applied techniques. Finally, it is recommended to incorporate these methods in WWTPs instead of using traditional methods to enhance toxic material removal and minimize the accumulation of antibiotics in our environment.

Conflict of Interest
There is no conflict of interest.
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