



## Review

## Removal of organic pollutants from aqueous solution using agricultural wastes: A review

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## ABSTRACT

Among various wastewater treatment techniques, adsorption is supposed as one of the best methods due to its inexpensiveness, universal nature and ease of operation. Then, the use of agricultural waste for organic pollutants adsorption from aqueous solution has been reviewed. This article focuses on the preparation of activated carbon through various agricultural wastes and their modified. Moreover, the adsorption capacity for organic pollutants (such as dyes, petroleum hydrocarbons, pharmaceuticals, pesticides, and other organics) and desorption approaches have been investigated. In addition, optimization of activated carbon preparation conditions and adsorption process variables using response surface methodology (RSM) was also summarized. Adsorption mechanisms of organic pollutants are briefly discussed. The review implied the potential of agricultural wastes for organic pollutants removal from wastewater.

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## Contents

1. Introduction . . . . .	740
2. Preparation of adsorbent by agricultural waste . . . . .	740
2.1. Composition of agricultural waste . . . . .	740
2.2. Preparation of AC and agricultural waste modification . . . . .	740
2.2.1. Preparation of AC by agricultural waste . . . . .	740
2.2.2. Agricultural waste modification . . . . .	742
3. Removal of organic pollutants . . . . .	744
3.1. Dye . . . . .	744
3.1.1. Cationic dye . . . . .	744
3.1.2. Anionic dye . . . . .	745
3.1.3. Nonionic dyes . . . . .	750
3.2. Petroleum hydrocarbons . . . . .	750
3.3. Pharmaceuticals . . . . .	751
3.3.1. Anti-inflammatory . . . . .	751
3.3.2. Antibiotics . . . . .	751
3.3.3. Other drugs . . . . .	752
3.4. Pesticides . . . . .	752
3.5. Other organics . . . . .	752
4. Optimization of AC preparation conditions and adsorption process variables by RSM method . . . . .	753
5. Regeneration of adsorbent . . . . .	755
6. Mechanism of adsorption . . . . .	756
7. Conclusion and future perspectives . . . . .	757

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Nomenclature . . . . .	758
Acknowledgments . . . . .	758
References . . . . .	

## 1. Introduction

Environmental pollution has reached a stage in which it should be seriously examined. However, among the various types of pollution, water pollution has attracted the attention of some researchers. The main sources of water contamination include the industrialization (textile, rubber, leather, paper, plastics, coal, food, petrochemical, pharmaceutical, dye industries, etc.), agricultural activities (the use of pesticides in agriculture, forestry, as well as veterinary and aquaculture drugs), municipal wastewater, other environmental and global changes [1–3]. A few hundred organic pollutants have been found to contaminate water resources [4]. These contaminations are very dangerous due to their various side effects and embryotoxicity, mutagenicity, teratogenicity, and carcinogenicity as well as health disorders to human beings, such as the dysfunction of kidney, reproductive system, liver, brain, and central nervous system. [5–8]. For more information on their side effects and toxicity, see these literatures (such as Meyers [9], Köhle [10]) Hence, it is necessary to provide an effective treatment for organic wastewater.

Currently, several treatment technologies are employed to remove organic pollutants from wastewater which include flocculation, coagulation, biological oxidation, and sedimentation [5], photo-Fenton treatment, advanced oxidation processes (AOPs) [7], oxidation with chemical oxidants (ozone or hydrogen peroxide, etc.), photocatalytic oxidation/degradation [11,12], membrane processes [13], electrochemical oxidation/degradation [14,15], adsorption and combined methods. Despite the availability of the above mentioned processes for the removal of organic pollutants, the adsorption process still remains the best because of its universal nature, inexpensiveness, ease of operation [4], flexibility, insensitivity to toxic pollutants, as well as high efficiency and effectiveness [16,17]. Moreover, adsorption can also remove soluble and insoluble organic pollutants without the generation of hazardous by-products [4,14].

In the past, organic pollutants have been removed by different species of adsorbents, such as nanometer materials (nano-graphite/Fe<sub>3</sub>O<sub>4</sub> composite [18], Fe<sub>3</sub>C-multiwalled carbon nanotubes (MCNT) [19], alkali-activated MCNT [20], MCNT-COOH [21], Pt/poly(2-methoxyaniline)-MCNT [22], CNT-Al<sub>2</sub>O<sub>3</sub> [23], etc.); industrial wastes (waste tires [24], coal fly ash [25,26], dried activated sludge and fly ash [27], silica gel [28], polyurethane foam [29], etc.); sea materials (crosslinked chitosan beads [30], chitosan/surfactant [31], peat moss [32], brown seaweed biomass [33], *Spirogyra* sp. [34], shrimp shell [35], etc.); soil and ore materials (montmorillonite clay [36], Moroccan clays [37], red mud [38], calcined Lapindo volcanic mud [39], river sediment [40], acrisol and phaeozem [41], hematite [42], halloysite–magnetite-based composite [43], kaolinite [44,45], magnetite, quartz [45], zeolite [46–48], bentonite [49], etc.); metal oxides and hydroxides (ZnCr- and MgAl-layer double hydroxide [50], TiO<sub>2</sub> [51], Mg–Fe layer double hydroxides [52], Fe<sub>3</sub>O<sub>4</sub>/ZnCr-layered double hydroxide composite [53], Fe<sub>3</sub>O<sub>4</sub>/MgAl layer double hydroxides [54], magnesium hydroxide [55], etc.), metal–organic frameworks (MOF) (Cr-based MOFs [56], iron terephthalate [57], MIL-101–Cr [58], Cu-BTC [59], Cu-BTC and Fe-BTC [60], etc.) agricultural wastes (rice husk [61–64], sugarcane bagasse [65–67], peanut husk [68, 69], sesame hull [70], tomato [71]), etc. However, agricultural waste adsorbents prepared have certain advantages such as renewable organic resource, abundant, low cost, selective adsorption effluent and easy regeneration [72].

In order to provide a summary of recent information concerning the use of agricultural wastes for preparation sorbents to adsorb organic

pollutants, a literature survey about 230 recently published papers were done and a review was written. However, in this area (removal of organic pollutants using low-cost adsorbents), a few reviews have also been written. For example, the removal of dye contaminants from wastewater by AC based on agricultural waste has been investigated [73]. Later, the use of low-cost adsorbents for the methylene blue (MB) removal has been reviewed [74]. Ali et al. [4] discussed low cost adsorbents which have wide range of applications in the removal of various sorts of organic pollutants. In addition, Salleh [75], Rangabhashiyam [76] and Anastopoulos [77] reviewed studies of dye adsorptions on agricultural wastes. Recently, a paper provides a comprehensive review on using typical low-cost biosorbents obtained from lignocellulose and chitin/chitosan for specific organic pollutants (such as phenolic compounds, PAHs, organic pesticides, and organic herbicides) adsorption [78]. But up to now, nobody focuses especially on using agricultural waste adsorbents prepared for the removal of organic pollutants (such as dyes, petroleum hydrocarbons, pharmaceuticals, pesticides, and other organics). In the study, the removal of organic pollutants onto agricultural waste adsorbents (raw material, modified agricultural waste or preparation AC by them) has been reviewed. Moreover, the adsorption capacities of organic pollutants desorption approaches, adsorption mechanism, and the optimizations activated carbon preparation conditions and adsorption process variables by response surface method have been summarized.

## 2. Preparation of adsorbent by agricultural waste

### 2.1. Composition of agricultural waste

Agricultural wastes are lignocellulosic materials which consist of three main structural components (lignin, cellulose and hemicelluloses) [75]. Lignin is the polymers of aromatic compound, and it has a complex three-dimensional structure. The basic chemical phenylpropane units of lignin are bonded together through a set of linkages to form a very complex matrix, which consist of various functional groups (such as hydroxyl, methoxyl and carbonyl). Moreover, hemicellulose is largely soluble in alkali and more easily hydrolyzed. As such, both hemicellulose and cellulose also contain of oxygen functional groups which are presented in lignocellulosic materials including hydroxyl, ether, and carbonyl [79]. These functional groups play an important role in the preparation process of adsorbents.

### 2.2. Preparation of AC and agricultural waste modification

#### 2.2.1. Preparation of AC by agricultural waste

In the past, some researchers have used different raw materials for the preparation of activated carbon (AC), such as plastic wastes (polyethylene terephthalate (PET) and polyvinyl chloride (PVC)), various industrial wastes (like fly ashes, pitch, and polymeric residues from factories), and other wastes (tires, sewage sludge, etc.) [80]. Essentially, it is one of the most important challenges to prepare AC with high cost for commercial manufacturers. However, in recent years, some groups have employed agricultural wastes to synthesize AC because they have high carbon content, low levels of inorganic compounds, considerable mechanical strength, low ash content and inexpensive cost. In particular, Ioannidou and Zabaniotou [81] have summarized the preparation of AC using agricultural residues and have reviewed the effects of various process parameters on the pyrolysis stage. Moreover, reaction kinetic modeling and activation of the pyrolytic char were also discussed. In general, the production of AC principally involves two

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