Review

Application of Magnetic Composites in Removal of Tetracycline through Adsorption and Advanced Oxidation Processes (AOPs): A Review

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Abstract: Water pollution induced by the tetracycline (TC) has caused global increasing attention owing to its extensive use, environmental persistence, and potential harm for human health. Adsorption and advanced oxidation processes (AOPs) have been promising techniques for TC removal due to ideal effectiveness and efficiency. Magnetic composites (MCs) which exploit the combined advantages of nano scale, alternative sources, easy preparation, and separation from wastewater are widely used for catalysis and adsorption. Herein, we intensively reviewed the available literature in order to provide comprehensive insight into the applications and mechanisms of MCs for removal of TC by adsorption and AOPs. The synthesis methods of MCs, the TC adsorption, and removal mechanisms are fully discussed. MCs serve as efficient adsorbents and photocatalysts with superior performance of photocatalytic performance in TC degradation. In addition, the TC can be effectively decomposed by the Fenton-based and SO4•− mediated oxidation under catalysis of the reported MCs with excellent catalytic performance. Based on the existing literature, we further discuss the challenge and future perspectives in MCs-based adsorption and AOPs in removing TC.

Keywords: magnetic composites; tetracycline; adsorption; advanced oxidation processes

1. Introduction

Antibiotics are widely used in industries such as medicine, animal husbandry, and aquaculture to kill various kinds of pathogenic bacteria [1,2]. Now, antibiotics considered as emerging environmental pollutants have received more attention in the world due to their chronic undesirable effect on the health of human beings and aquatic biota [3,4]. Tetracycline (TC) is one of the main antibiotics groups used for veterinary purposes, human therapy, and agricultural purposes [5,6]. TC was the most frequently used antibiotics and widely distributed in the aquatic environment [7,8]. However, for humans or animals, it is difficult to metabolize TC, and most of the TC is excreted in the form of original drug or parent compounds in the environment [9,10]. As a result, more than 50% of the TC enter the environment as metabolites [1,11]. Even humans and animals can excrete 50–80% of the administered dose of TC as the parent compound through urine [7]. Moreover, TC has been detected widely in different water environments: mariculture
(0.2–259.1 ng L\(^{-1}\)) [12], drinking water sources (11.16 ng L\(^{-1}\)) [13], and groundwater (0.4 ug L\(^{-1}\)) [14]. Long-term residual TC may result in the proliferation and transmission of drug-resistant bacterial flora, which may in turn affect the structure and function of ecosystems [15]. The long-term harmful effects of residual TC on human health and ecosystems have become a serious concern [1]. For example, the emergence of these antibiotic residues inhibits the growth and development of aquatic species, and may over-accumulate in the food chain to lead to joint disease, nephropathy, endocrine disruption, and central nervous system defect [16,17]. The techniques for removing TC from water include biological processes [18], coagulation [3], sedimentation [19], electrochemical processes [20], membrane techniques [21], advanced oxidation processes [22], chlorination [23], and adsorption [4], which are summarized in Figure 1. However, most of conventional treatment processes have an inherent disadvantage for removing TC [24]. For example, the chlorination method could produce intermediate products with higher toxicity [25], membrane techniques did not actually remove or degrade the TC, but only transferred it to a new phase, which could cause secondary contamination [26], and the mass transfer efficiency of electrochemical oxidation on metal electrodes was restricted for practical application [27,28]. Among these, adsorption and advanced oxidation processes were relative cost-effective and efficient methods. Adsorption is considered to be one of the most promising methods to remove TC from aqueous solutions due to its attractive advantages such as low-cost effectiveness, environmental friendliness, and convenient operation [29,30]. Ahamed et al. [28] prepared magnetic nanocomposites with a high surface area of 376 m\(^2\) g\(^{-1}\), high pore volume of 0.38 cm\(^3\) g\(^{-1}\), and the adsorption capacity of 215.3 mg g\(^{-1}\) for TC. Advanced oxidation processes (AOPs) (photocatalysis, electrochemical oxidation method, Fenton-like method, etc.) are widely used to remove TC due to high efficiency, cost-effectiveness, and environmental friendliness [31–33]. Sun et al. [34] found that the N-deficient g-C\(_3\)N\(_4\)/PS (g-CN\(_x\)/PS) system displayed a high efficiency of TC degradation under photocatalysis with over 80% after three recycles, indicating that the carbon nitride based photocatalyst possessed excellent photocatalytic stability. Numerous catalysts are reported to be utilized in AOPs, such as TiO\(_2\) [35], WO\(_3\) [36], Fe\(_2\)O\(_3\) [37], ZnO [38], Ag\(_3\)PO\(_4\) [39], graphene [40], SnO\(_2\) [41], etc. However, most of these catalysts were hard to be separated from water [42,43] and tended to agglomerate [37]. Under these circumstances, magnetic materials have garnered considerable interest as they can overcome the above shortcomings of normal catalyst [44] and exhibit the enhanced degradation efficiency via the synergistic effect of the combination of the host and guest compounds [45].

![Current remediation technologies for TC](image)

**Figure 1.** Overview of existing remediation technologies for controlling the TC pollution.

Magnetic composites (MCs) (such as magnetic biochar, magnetic nanomaterials, magnetic chitosan, etc.) are broadly utilized in adsorption and AOPs due to their high surface area, porous structure, convenient separation, and recycling [1,46–48]. Transition metal
salts, natural iron ores, and iron oxides were commonly used magnetic precursor. When the loaded magnetic species have strong magnetism, the magnetic performance of MCs will be relatively superior [49]. For example, Li et al. [50] prepared magnetic biochar composites by pyrolysis of siderite and rice husk and the presence of specific saturation magnetization was 9.45 emu/g. Sherlala et al. [51] found that the chitosan-magnetic-graphene oxide (CMGO) nanocomposite had an excellent saturation magnetization (49.30 emu/g), which could be easily separated from the solution by the application of an external magnetic field. The methods of MCs production include hydrothermal [52], coprecipitation [37] and sol–gel methods [53], etc. In general, the pure magnetism has the disadvantages of bad dispersion, poor separation effect, weak electron transfer ability, and low catalytic activity. To overcome the above disadvantages, researchers used different precursors and original magnets to prepare MCs, most of which were fictionalizing by porous or photoreceptive particles, such as activated carbon [37], graphitic carbon nitride [24], and titanion oxide [54], to enhance their feasibility for TC removal from wastewater. In some cases, original magnets were combined with porous supporters with high specific surface area to address the problem that magnets had a strong tendency to agglomerate [37]. In other cases, non-magnetic catalyst could be coupled with magnets to facilitate their recycling efficiency [52]. In terms of TC removal from wastewater, even though diverse MCs were developed for TC removal from wastewater, there was no a systematic review on the fabrication of MCs and the mechanisms of their application in TC removal.

To date, applications of MCs in water treatment has aroused considerably the interest of researchers, as several previous reviews on MCs have been published in the past few years [24,49,55–57]. For example, Li et al. [58] only focused on the synthesis and environmental remediation of magnetic biochar and Minile et al. [24] only summarized graphene-based materials to remove TC in aqueous solution by photocatalytic degradation and adsorption. The above reviews only focused on MCs based on single matrix, or a certain AOPs technique, but no review systematically introduced the degradation of a pollutant by various MCs via AOPs.

In light of this, we focused more attention on the applications and mechanisms of MCs for TC removal through adsorption and AOPs based published literature by summarizing the reported MCs for TC removal by absorption and AOPs. Firstly, we discuss the preparation methods of magnetic materials and categorizing the MCs. Secondly, we introduce the possible removal mechanisms between TC and MCs by adsorption and AOPs degradation. Thirdly, we investigate the synergistic effect between components during the degradation process. Finally, the possible challenges and outlook to appreciate more prospective improvements in similar future efforts are presented. This review may fulfill the existing knowledge gaps and provide favorable suggestions in TC removal from wastewater for future studies.

2. Different Kinds of MCs and Their Fabrication Methods

Previous reviews have summarized in details the application and development prospects of MCs in various fields. Brião et al. [59] introduced chitosan-based magnetic adsorbents to remove toxic heavy metals. Jacinto et al. [60] focused on the main synthesis processes of magnetic photocatalysts, and their effect on the catalyst morphology, degradation efficiency, and recycling. In this review, we briefly introduce the categories and synthesis methods of MCs for TC removal.

2.1. Types of Magnetic Materials

The precursors of MCs have a wide range of sources, such as biochar, activated carbon, chitosan, cellulose and artificial polymer. For example, Bao et al. [61] used the coprecipitation method to successfully synthesize magnetic illite clay-composite material (FeO@illite). Bai et al. [62] modified copper ferrite on the surface of molybdenum disulfide to prepare MoS2/CuFe2O4 nanocomposites. According to the classification method of MCs such as magnetic biochar and chitosan-based MCs, and combining the precursors of MCs,
this review would divide MCs into four categories: carbon-based MCs, polymer-based MCs, metal–organic framework (MOFs) based MCs, and others. The specific types of MCs are shown in Figure 2.

![Categories of magnetic composites.](image)

**Figure 2.** Categories of magnetic composites.

### 2.2. Synthesis Methods for MCs

There are many common preparation methods for MCs, including: pyrolysis, co-precipitation, hydrothermal/solgel, sonochemica, impregnation, post-crosslinking, amination, polymerization, in-situ precipitation, and oxidation. Diverse preparation methods for MCs in reported articles for TC removal are summarized in Table 1.

**Table 1.** The main preparation methods for magnetic materials.

| Material Category               | Species                          | Magnetic Materials                                      | Synthesis Techniques          | References |
|--------------------------------|----------------------------------|--------------------------------------------------------|-------------------------------|------------|
| **Carbon-based magnetic materials** | **Graphene**                      | Sodium citrate coated Fe$_3$O$_4$ nanoparticles         | Pyrolysis, co-precipitation   | [54]       |
|                                 | **Graphene**                      | Thiourea-dioxide–reduced magnetic graphene oxide        | Pyrolysis, co-precipitation   | [63]       |
|                                 | **Graphene**                      | Nitrilotriacetic acid-functionalized magnetic graphene oxide | Pyrolysis, co-precipitation, Hydrothermal/Solgel | [64]   |
|                                 | **Graphene**                      | Magnetic graphene oxide/ZnO nanocomposites              | Pyrolysis, co-precipitation, Hydrothermal/Solgel | [65]       |
|                                 | **Biochar**                       | MnFe$_2$O$_4$/activated carbon magnetic composite       | Pyrolysis, co-precipitation   | [66]       |
|                                 | **Biochar**                       | Magnetic porous carbon from waste hydrochar             | Pyrolysis                     | [67]       |
|                                 | **Biochar**                       | Sugarcane bagasse magnetic carbon composites            | Pyrolysis                     | [68]       |
|                                 | **Biochar**                       | Activated sawdust hydrochar                             | Pyrolysis                     | [69]       |
|                                 | **Biochar**                       | Magnetic chicken bone biochar                            | Pyrolysis, co-precipitation   | [70]       |
|                                 | **Biochar**                       | Alkali-acid modified magnetic biochar                    | Pyrolysis, hydrothermal/solgel | [71]       |
|                                 | **Biochar**                       | Magnetic carbon-coated cobalt oxide nanoparticles       | Sonochemical, pyrolysis       | [72]       |
| Material Category | Species | Magnetic Materials | Synthesis Techniques | References |
|------------------|---------|--------------------|----------------------|------------|
| Biochar          | Modification and magnetization of rice straw derived biochar | Pyrolysis, impregnation method | [47] |
| Biochar          | Ferroferric oxide nanoparticles assisted powdered activated carbon | Co-precipitation | [30] |
| Biochar          | Biochar-supported iron-copper bimetallic composite activating oxygen system | Pyrolysis, co-precipitation | [73] |
|                  | Hydrothermal synthesis of magnetic sludge biochar | Pyrolysis, Hydrothermal/solgel | [74] |
|                  | Carbon disulfide-modified magnetic ion-imprinted chitosan-Fe(III) | Co-precipitation, Hydrothermal/solgel, copolymerization, post-crosslinking, and amination | [69] |
| Resin            | Novel magnetic multi-amine resins | Co-precipitation | [75] |
| Urethane polymer | Sustainable magnetic polyurethane polymer nanocomposite | Co-precipitation | [76] |
|                  | Chitosan based magnetic nanocomposite | Copolymerization, sonochemical, hydrothermal/solgel | [46] |
|                  | NiFe₂O₄-COF-chitosan-terephthalaldehyde nanocomposites film | Sonochemical | [77] |
| Resin            | magnetic multi-amine decorated resin | Co-precipitation, polymerization, post-crosslinking reactions, and amination. | [5] |
| MOFs             | MOFs-chitosan composite beads | Hydrothermal reaction or solvothermal reaction | [78] |
|                  | Fe-based MOFs | Solvothermal method | [79] |
| Others           | Magnetic adsorbent constructed from the loading of amino functionalized Fe₃O₄ | Solvothermal method | [80] |
|                  | La-modified magnetic composite | Co-precipitation | [81] |
|                  | Co-existing TiO₂ nanoparticles magnetically modified kaolin | In-situ precipitation and oxidation | [82] |

Carbon-based MCs were mainly prepared by pyrolysis and co-precipitation. Pyrolysis was the main method to produce carbon-based magnetic composites, while co-precipitation had the advantages of scalable, simple, and easy size/morphology control. For example, Yu et al. [54] and Yang et al. [63] prepared thiourea dioxide reduced magnetic graphene oxide by pyrolysis and co-precipitation. In addition, polymer-based MCs were often prepared by various methods, such as co-precipitation, hydrothermal/solgel, sonochemical, and so on. The materials prepared by sonochemical had high efficiency and fine particle size, and hydrothermal/solgel had the advantages of high yield, water-based medium, scalability, and energy saving. For MOFs-based MCs, pyrolysis was the main method to prepare MOFs-based MCs. For example, Xiao et al. [2] prepared Nico/Fe₃O₄-MOF-74 composite by pyrolysis. Besides, there are other preparation methods to be used. For example, Zhao et al. [78] prepared MOF–chitosan composite beads by hydrothermal reaction or solvothermal reaction.

There are also some magnetic composites that do not belong to the above three types. Their preparation methods could change greatly with the different research materials. For example, Wang et al. [83] and others prepared co-existing TiO₂ nanoparticles magnetically modified kaolin by in-situ precision. Mi et al. [81] prepared La-modified magnetic composite by co-precipitation.
### Applications of MCs for TC Adsorption

Up to date, many research articles have focused on applications of MCs for TC removal. Previous studies on the application of MCs for TC removal by adsorption in aqueous solution are summarized in Table 2.

#### Table 2. Performance of diverse magnetic materials for TC adsorption.

| Material                                                   | Initial Concentration of TC (mg/L) | Dosage (g/L) | pH | Adsorption Conditions | Adsorption Capacity (mg/g) | Isotherms/Kinetics Model                  | References |
|------------------------------------------------------------|------------------------------------|--------------|----|-----------------------|----------------------------|------------------------------------------|------------|
| Fe₃O₄ magnetized graphene oxide sponge                     | 400                                | 0.625        | 3  | 308 2880             | 473                        | Temkin model/pseudo-second-order model   | [54]       |
| Ferromanganese oxide magnetically bonded biocarbon        | 100                                | 0.4          | 6  | 318 1440             | 101                       | Freundlich model/pseudo-second-order model | [27]       |
| Magnetic nano-scale biosorbent Fe₃O₄ magnetized porous carbon | 10                                  | -            | 6  | 303 -                 | 56.0                      | Langmuir model/pseudo-second-order model | [84]       |
| Fe₃O₄/activated carbon                                    | 30                                 | 1            | -  | 303 7200             | -                         | Freundlich model/pseudo-second-order model | [67]       |
| MnFe₂O₄/activated carbon                                  | 222                                | 1            | 5  | 298 -                 | 591                       | Freundlich model/pseudo-second-order model | [66]       |
| Fe₃O₄ magnetized chicken bone biochar                     | 100                                | 10           | 8  | 299 1440             | 93.2                      | Freundlich model/pseudo-second-order model | [70]       |
| Nitrilotriacetic acid-functionalized Fe₃O₄ magnetized graphene oxide: | 50                                 | 0.192        | 4.0| 298 1440             | 212                       | Langmuir model/pseudo-second-order model | [85]       |
| Magnetic hydrochar                                        | 100                                | 0.4          | -  | 298 120              | 424                       | Langmuir model/pseudo-second-order model | [86]       |
| Thiourea dioxide–reduced Fe₃O₄ magnetized graphene oxide   | 10                                 | 70           | 4  | 313 1440             | 1233                      | Langmuir model/pseudo-second-order model | [63]       |
| Modified Fe₃O₄ magnetized polyoxometalates nanoparticle   | 150                                | 1            | 6.8| 298 1440             | 133                       | Temkin model/pseudo-second-order         | [80]       |
| Fe₃O₄@ZIF-8 microspheres                                  | -                                  | 2.5          | -  | 318 120              | 402                       | Langmuir model/pseudo-second-order kinetics model | [87]       |
| Carbon disulfide-modified magnetic ion-imprinted chitosan–Fe (II) | 100                                 | 0.5          | 8  | 298 -                 | 516                       | Langmuir model/pseudo-second-order model | [69]       |
| Fe₂O₃/Fe₃O₄ nanoporous activated carbon composite          | 150                                | 0.1          | 4  | 323 270              | 60.6                      | Langmuir model/pseudo-second-order model | [88]       |
| Fe₃O₄ magnetized starch polyurethane                      | 20                                 | 2.5          | 6  | 298 240              | 16.4                      | Freundlich and Redlich–Peterson isotherm models/pseudo-4th order model | [76]       |
| Fe₃O₄ magnetized resin                                    | 100                                | 0.2          | -  | 303 -                 | -                         | Freundlich model/pseudo-second-order model | [89]       |
| Fe₃O₄ magnetized imprinted polymer nanoshell              | 88.89                              | 0.5          | -  | 298 720              | 55.0                      | Langmuir model/pseudo-second-order model | [90]       |
| Fe₃O₄ magnetized carbon composites                        | 80                                 | 2            | 6.8| 303 1560             | 48.4                      | Freundlich model/pseudo-second-order model | [68]       |
| Fe₃O₄ magnetized polystyrene EDTA microsphere             | 40                                 | 3            | 6.3| 303 720              | 166                       | Temkin model/pseudo-second-order model   | [91]       |
| Fe₃O₄ magnetized macro-reticulated cross-linked chitosan   | -                                  | 2            | -  | -                    | 120                       | Freundlich model/pseudo-second-order model | [92]       |
| Auricularia-based Ni nanoparticles Fe₃O₄ magnetized porous carbon | -                                  | -            | -  | 318 720              | 397                       | Langmuir model/pseudo-second-order model | [93]       |
| Fe₃O₄ magnetized chitosan nanoparticles                    | 50                                 | 0.5          | 5.0| 298 2880             | 78.1                      | Langmuir model/pseudo-second-order model | [94]       |
| Zr(VI)-based metal organic framework Uio-66-(COOH)₂/GO | 25                                 | 0.4          | 7  | 298 1440             | 146                       | Freundlich model/pseudo-second-order model | [81]       |
| Alkali-acid modified magnetic biochar                     | 100                                | 0.5          | -  | 298 2880             | 165                       | Langmuir model/pseudo-secondary kinetic model | [95]       |
| Magnetic carbon-coated cobalt oxide nanoparticles (CoOBC) | 200                                | 1            | 7  | 318 1440             | 172.0                     | Langmuir–Freundlich model/pseudo-second-order kinetics | [71]       |
| Chitosan based magnetic nanocomposite                     | 700                                | 2.5          | 5  | 298 20               | 81.5                      | Langmuir model/pseudo-second-order model | [96]       |

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**References:**
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Table 2. Cont.

| Material | Initial Concentration of TC (mg/L) | Dosage (g/L) | Adsorption Conditions | Adsorption Capacity (mg/g) | Isotherms/Kinetics Model | References |
|----------|----------------------------------|-------------|----------------------|--------------------------|-------------------------|------------|
| Magnetic cellulose | 100 | 1 | 7 | 298 | 2880 | 44.9 | Freundlich model /Weber–Morris curve | [97] |
| Metal–organic framework MIL-101(Cr) loaded nano zero-valent iron | 100 | 0.15 | - | 318 | 120 | 625 | Langmuir model /pseudosecond-order model | [98] |
| Magnetic Fe/porous carbon hybrid (MagFePC) | 140 | 0.05 | 7 | 298 | 1440 | 1301 | Langmuir model /pseudo-second-order model | [99] |
| Magnetic chicken bone biochar (MCB) | 100 | 1 | 8 | 299 | 1440 | 98.9 | Freundlich isotherm | [70] |
| Fe₃O₄-g-CN@PEI-β CD NC | 265 | 0.04 | 9.2 | 320.1 | 20 | 833 | Langmuir model /pseudo-second-order model | [100] |
| Magnetic sludge biochar (Fe/Zn-SBC) | 200 | 0.2 | - | 298 | 1440 | 145 | /pseudo-second-order model | [74] |
| NiFe₂O₄-COF-terephthalaldehyde nanocomposites film (NCCT) | 100 | 0.17 | 8 | - | 2400 | 389 | /pseudo-second-order model | [77] |
| Magnetic graphene oxide/ZnO nanocomposites (MZ) | 500 | 0.278 | 6 | - | 2400 | 1590 | /pseudo-second-order kinetics model | [65] |
| Fe-based metal-organic frameworks | 100 | 4 | - | 298 | 1440 | 421 | /pseudo-second-order kinetics model | [79] |

3.1. Carbon-Based MCs

3.1.1. Graphene-Based MCs

Graphene have gained considerable interests among many researchers due to high mechanical strength and chemical stability [24]. Moreover, Graphene is used as the most effective TC adsorbent due to its large surface area and tunable structure. Well-designed structural modifications of 2D graphene with three-dimensional (3D) substrates, such as metal ions and their oxides, bio-molecules, and hydrogels, offers outstanding platform for adsorption [101,102]. However, the original graphene is rarely used for TC, because it is not easy to react with organic matter [103,104]. Thus, researchers use chemical modification to enhance its adsorption capacity and promote its application [104]. Graphene oxide (GO) and reduced graphene oxide (rGO) are widely used derivatives of graphene, both in their pristine and composite form, in the fields of adsorption [24,105]. The adsorption effect of graphene oxide and reduced graphene oxide materials on TC will be specifically discussed in the section.

Magnetic graphene composites were prepared to facilitate the separation of graphene adsorbents. The researches have shown that magnetic graphene oxide sponge (MGOS) prepared by freeze-drying nano-Fe₃O₄ particles with well graphene oxide (GO) dispersion could effectively adsorb TC with adsorption capacity of 473.0 mg g⁻¹, which was 50% higher than that of GO [54]. Moreover, the adsorption process was fast, and the pH and ionic strength had little effect on the adsorption. In addition, Li et al. [85] grafted nitrotriacetic acid onto magnetic graphene oxide (NDMGO) to adsorb TC from water and hydrogen bonds, amidation reaction, π–π, and cation–π interaction were the adsorption mechanism between NDMGO and TC. Yang et al. [63] used magnetic graphene oxide (TDMGO) to remove TC from aqueous solution. The maximum adsorption capacity of TDMGO for TC was 1233.0 mg g⁻¹ and the pH had little effect on adsorption. The pseudo-second-order kinetic model and Langmuir isotherm provided the better correlation for the experiment data. In addition, Qiao et al. [65] found that magnetic graphene oxide/zinc oxide nanocomposite (MZ) showed an excellent adsorption capacity for TC with adsorption capacity of 1590.3 mg g⁻¹ and could be easily recycled. Electrostatic attraction, π–π interaction, hydrogen bond, and cation exchange and complexation were the main modes of action. Shan et al. [106] 3D prepared reduced graphene oxide/nano-Fe₃O₄ hybrid hydrogel (3D-rGO/Fe₃O₄) to remove TC from aqueous solution. The 3D-rGO/Fe₃O₄ could effectively adsorb TC with adsorption capacity of 2044.4 mg g⁻¹. Bao et al. [107] prepared manganese ferrite–rGO (MnFe₂O₄/rGO) composite for TC removal with adsorption ca-
pacity of 1131.0 mg g$^{-1}$. The growth of MnFe$_2$O$_4$ played an important role to enhance TC removal.

### 3.1.2. Biochar-Based MCs

Biochar was used for environmental remediation and received extensive attention in regards to the removal of organic pollutants in water [58,108,109]. To achieve an easy separation in application, magnetic biochar has been extensively studied. Shao et al. [66] found that MnFe$_2$O$_4$/activated carbon magnetic composites had excellent performance for TC in aqueous solution. The results indicated that the adsorption capacity was 261.8 mg g$^{-1}$, and accorded the pseudo-second-order kinetic model. Song et al. [96] prepared hybrid nanocomposites of zero-valent iron loaded the activated carbon (ZVI@ACCS) to adsorb TC and the synergistic interactions of the electrostatic attraction, the bridging complexation, and the surface complexation could be used to explain the mechanism of adsorption. The biochar-supported iron–copper bimetallic composites (BC-FeCu) were successfully prepared by Liu et al. [73]. The adsorption and degradation of TC by BC-FeCu accounted for 26.1% and 73.9% of the total removal rate, respectively. Yang et al. [72] prepared magnetic carbon-coated cobalt oxide nanoparticles (CoO@C) to remove TC with an adsorption capacity of 769.4 mg g$^{-1}$. Zeta-potential and X-ray Photoelectron Spectroscopy analysis showed that there was a strong electrostatic interaction between the positive charge on the surface of CoO@C and TC.

The texture of biochar would affect the adsorption capacity of TC on the MCs. Dai et al. [47] modified rice straw biochar by an alkali–acid combined magnetization method. The adsorption capacity could reach 98.3 mg g$^{-1}$, and the main adsorption mechanisms were the hydrogen bonding and pore filling effect. Ma et al. [74] used municipal sludge biochar to synthesis magnetic sludge biochar (Fe/Zn-SBC) for removing TC. The results showed that the maximum adsorption capacity of Fe/Zn-SBC was 145.0 mg g$^{-1}$ and the adsorption process was dominated by pore filling, complexation of oxygen-containing groups, π–π conjugation, and hydrogen bonding. Rattanachueskul et al. [68] transformed bagasse into a new type of magnetic carbon composite and the maximum adsorption capacity was 48.4 mg g$^{-1}$. The adsorption of TC by magnetic adsorbents was mainly realized by the interaction between hydrogen bond and TC.

### 3.2. Polymer-Based MCs

#### 3.2.1. Chitosan-Based MCs

Polymer nanocomposites have been used to remove organic and inorganic pollutants from aqueous solutions [59]. Among all kinds of natural polymers, chitosan is the second most abundant natural biopolymer [69]. Chitosan has been considered as a promising adsorbent because of the existence of amino and hydroxyl groups. Chen et al. [69] reported magnetic ion imprinted chitosan-Fe (III) composite have good adsorption capacity of 516.3 mg g$^{-1}$. Li et al. [77] found that NiFe$_2$O$_4$-COF–chitosan–terephthalaldehyde nanocomposite film (NCCT) was an effective adsorbent for TC with the adsorption capacity of 388.5 mg g$^{-1}$. Complexation, cation exchange, electrostatic attraction, hydrogen bond, and π–π interaction were the adsorption mechanisms of TC on NCCT. Ahamad et al. [28] used MCs prepared by chitosan, thioarbituric acid, malondialdehyde, and Fe$_3$O$_4$ nanoparticles (CTM@Fe$_3$O$_4$) to absorb TC with an adsorption capacity of 215.3 mg g$^{-1}$. Langmuir model and the pseudo-second-order nonlinear model were the best models for fitting adsorption isotherms and adsorption kinetics.

#### 3.2.2. Resin-Based MCs

Amino rich resin has attracted the attention of researchers in recent years due to its excellent adsorption capacity, good affinity, and chemical stability [2]. For example, Zhu et al. [75] prepared several magnetic polyamine resins (MMARs) for the removal of TC which showed that the specific surface area could reach up 1433.4 m$^2$ g$^{-1}$ and their adsorption capacity for TC reached 107.9 mg g$^{-1}$. Wang et al. [5] observed that
magnetic polyamine modified resin (MMAR-G) could be used to adsorb TC with the adsorption capacity of 46.2 mg g\(^{-1}\). In short, the behavior of TC adsorption on polymer-based magnetic composites generally followed the pseudo-second-order kinetic model and Langmuir model.

### 3.3. Metal–Organic Framework (MOFs)

MOF was a new type of highly ordered porous crystal hybrid materials with infinite skeleton structure, which can be self-assembled by multi-functional organic ligands and metal centers. Compared with traditional porous materials, MOFs have high porosity, high specific surface area, and rich active functional groups. They have attracted much attention because of their unique performance.

Xiao et al. [2] found that NiCo/Fe\(_3\)O\(_4\)-MOF-74 magnetic composites had good enrichment and removal ability for TC, and the removal rate reached 94.1% in 5 min. The main interaction between adsorbents and TC was likely to have more available metal sites that could form stable metal ligands with TC. Zhang et al. [79] used three kinds of iron-based MOFs with different pore properties and open metal centers to remove tetracycline hydrochloride (TCH). Among them, MIL-101 (Fe) showed excellent adsorption performance for TCH with adsorption capacity of 420.6 mg g\(^{-1}\) due to much open metal centers and higher binding energy. In addition, the MOFs were uniformly and stably immobilized in the chitosan matrix [78]. The results showed that the maximum adsorption capacity could reach 495.0 mg g\(^{-1}\) and the pseudo-second-order model and Langmuir isotherm model could fit the adsorption process. The adsorption mechanisms included electrostatic interaction, π–π stacking interaction, and hydrogen bond interaction. Gu et al. [99] also found that TC was removed efficiently by synergistic adsorption of carbon and iron. The adsorption capacity could reach up 1301.2 mg g\(^{-1}\) when the pH was about 7.

### 3.4. Others

In addition to the above three types of MCs, there are other types of MCs with unique properties and advantages in adsorbing TC. Mi et al. [81] introduced element lanthanum into magnetic substrate to improve its adsorption performance. The Langmuir model fitting results showed that the adsorption of TC on MCs could reach 145.9 mg g\(^{-1}\). Ou et al. [80] synthesized magnetic polyoxometalate adsorbents and found that there was a strong hydrogen bond between NH\(_2\)-Fe\(_3\)O\(_4\) and CC/POMNP, which kept the stability of the adsorbent. Wang et al. [83] studied the effect of nanometer titanium dioxide (TiO\(_2\)) on the adsorption and desorption of TC by magnetized kaolin (MK). The results showed that TiO\(_2\) nanoparticles increased the adsorption capacity of TC on MK by 2.02%.

Generally, previous studies have demonstrated that MCs have exceptional performance in removing TC. To date, carbon-based MCs, polymer-based MCs, and MOFs have been mainly used to remove TC in aqueous solution. Carbon-based MCs and polymer-based MCs were the most widely used because of the porous structure and the huge specific surface area. Moreover, magnetic graphene and metal-modified magnetic biochar showed excellent performance in removing TC from aqueous solutions. Carbon-based MCs and graphene-based MCs could effectively remove TC by electrostatic, π–π EDA, cation–π bonding, hydrogen bonding, and hydrophobic interactions. Polymer-based MCs were mainly realized by complexation, cation exchange, electrostatic attraction, hydrogen bonding, and π–π interaction to adsorb TC. The mechanisms of MOFs–MCs adsorbing TC mainly included electrostatic interaction, π–π stacking interaction, and hydrogen bonding interaction because of their high porosity, large specific surface area, and abundant active functional groups.

### 4. Magnetic Composites-Catalyzed Advanced Oxidation Processes (AOPs)

In recent decades, AOPs have been considered as the most effective methods for organic pollutants degradation in water because the generated active oxygen species can destroy organic pollutants into innocuous or low-toxic small compounds [110,111].
Common AOPs include Fenton process, photocatalysis, microwave enhanced AOPs, electrochemical oxidation, and ultraviolet radiation.

MCs are widely used in photocatalysis, Fenton, and Fenton-like systems, not only in the degradation of antibiotics [112–114], but also in other organic pollutants, such as dye [115], pesticides [116], etc. With light activation or assistance by H$_2$O$_2$, persulfate, or other oxidants, MCs can generate active radicals such as HO$_2$•, HO•, SO$_4$•−, which induce a series of following reactions to decompose antibiotics [67,108,117]. In this section, the current development and application of MCs in degradation of TC through AOPs are systematically discussed. Specifically, the degradation of antibiotics by MCs could be divided into the following categories.

4.1. Hydrogen Peroxide Based Advanced Oxidation Processes (H-AOPs)

H-AOPs is the most frequently used AOPs in TC disposal due to its environmental friendliness [117]. Simply, catalysts will activate hydrogen peroxide (H$_2$O$_2$) under acidic conditions to generate highly reactive hydroxyl radicals. Various magnets or magnetic ferrites were employed to activate H$_2$O$_2$, such as Fe$_3$O$_4$ [112], CuFe$_2$O$_4$ [31] and ZnFe$_2$O$_4$ [118], etc. However, various magnets were of high density and less exposed active sites, which restrained the catalytic activity. Generally, the introduction of supports can overcome these limits and enhance efficiency of catalysts, and the simplified reactions in the photo-Fenton process were as follow [119]:

\[
\begin{align*}
\text{Fe}^{3+} + h\nu & \rightarrow \text{Fe}^{2+} + \text{HO}• + \text{H}^+ \\
\text{Fe}^{2+} + \text{H}_2\text{O}_2 & \rightarrow \text{Fe}^{3+} + \text{HO}• + \text{OH}^- 
\end{align*}
\]

Lai et al. [120] synthesized the MnFe$_2$O$_4$/biochar composite to degrade TC in photo-Fenton system, the existence of biochar made the MnFe$_2$O$_4$ more stable and improved the degradation efficiency, and this catalyst could achieve more than 90% removal rate within a pH range of 3 to 9. In the degradation process, TC was adsorbed onto the biochar surface, making it convenient for radicals to attack TC. Xin et al. [121] synthesized biochar modified CuFeO$_2$ as Fenton-like catalyst to degrade TC. The CuFeO$_2$/BC-1.0 have a strong reusability, higher catalytic activity and high stability due to the synergistic effect of Fe$^{3+}$/Fe$^{2+}$ and Cu$^{2+}$/Cu$^{+}$ redox cycles. Yu et al. [122] used Fe$_3$O$_4$-decorate hierarchical porous carbon skeleton (Fe$_3$O$_4$@MSC) to degrade TC. The TC degradation efficiency reached up 99.2% after 40 min. Kakavandi et al. [123] used Fe$_3$O$_4$ coated activated carbon (AC@Fe$_3$O$_4$) as a peroxidase mimetic to degrade TC through Fenton-like catalytic progress. AC@Fe$_3$O$_4$ had high activity and degradation efficiency after five concessive cycles.

Biopolymers, such as chitosan and alginate, are recognized as efficient supporters for catalytic applications. For example, Li et al. [124] observed that magnetite nanoparticles were successfully embedded into chitosan beads and used for efficiently degrading TC. The results showed that about 96.0% of TC was degraded within 20 min. The stable porous structure, abundant active sites, and possible synergistic effects between two components enhance the degradation performance of beads.

Recently, Fe-based MOFs have become a popular catalyst in which the Fe was proved to be a catalytically active center. Wu et al. [125] prepared Fe-based MOFs as Fenton-like catalysts for TC–HCl degradation. The result showed that the removal efficiency of TC–HCl and the apparent rate constant reached the maximum with adding 0.15 g L$^{-1}$ catalysts and 10 mL L$^{-1}$ H$_2$O$_2$. Fe-based MOFs exhibited the best Photo-Fenton performance mainly attributed to its largest surface area and pore volume, and the most coordinately unsaturated iron sites.

As magnetic adsorbents or magnetic catalysts, iron-based materials have excellent properties due to electrical and magnetic properties [126,127]. Many studies have confirmed that iron-based materials were important component magnetic mineral composites. For example, Semeraro et al. [128] prepared a composite catalyst based on zinc oxide (ZnO) and iron oxide (γ-Fe$_2$O$_3$) by a microwave-assisted aqueous solution method to degrade TC
in aqueous solution. The results clearly demonstrated that the ZnO/γ-Fe₂O₃ composite catalyst presented significant photocatalytic activity with the degradation efficiency of 88.5%. Lian et al. [129] observed that magnetic palygorskite nanoparticles (Pal@Fe₂O₃) could efficiently degrade TC in a wide pH range of 3–7. In addition, Chen et al. [130] prepared NiFe₂O₄/C yolk-shell nanostructure using polyacrylic acid sodium salt as a template. Notably, the degradation rate of TC reached 97.2% in 60 min under visible light irradiation (λ > 400 nm) with NiFe₂O₄/C. Qin et al. [131] investigated the catalytic activity of magnetic core–shell MnFe₂O₄@C and MnFe₂O₄@C-NH₂ in the antibiotic degradation. Compared to MnFe₂O₄, MnFe₂O₄@C and MnFe₂O₄@C-NH₂ presented higher catalytic activity in the antibiotics and TOC removal. Mashayekh-Salehi et al. [132] found that pyrite from mine waste was an excellent mineral catalyst and •OH was the main oxidizing species in a heterogeneous Fenton-like pyrite/H₂O₂ process. More than 85% of TC was removed in 60 min.

To sum up, different degradation mechanisms or processes during the MCs/Fenton systems were summarized as follows. Yu et al. [122] observed that the UV assisted heterogeneous Fenton-like process in Fe₃O₄@MSC improved the cycle of Fe³⁺/Fe²⁺ and activated the interfacial catalytic site. The •OH played an important role in the catalytic reaction. Li et al. [124] found that TC was adsorbed onto the beads’ surface from the bulk solution through π–π action between the benzenenings of TC and mesoporous Fe₂O₃-Cs beads. Moreover, the Fe₃O₄-Cs catalyst could catalyze H₂O₂ to form •OH to attack TC through intramolecular electron transfer process. Wu et al. [125] reported that Fe–MOFs were effective Fenton-like catalysts and •OH was the key reactive oxidative species. In addition, Nie et al. [133] found that •OH and •O₂⁻/•HO₂ were involved in TC degradation by using the Fe₃O₄-S/H₂O₂ system. Chen et al. [130] suggested that NiFe₂O₄/C was excited to generate electron–hole pairs in this conduction band and the valence band under visible-light irradiation, which reacted with Fe³⁺ to form Fe²⁺ to directly reacted with H₂O₂ to produce •OH. Simultaneously, the holes of NiFe₂O₄/C were directly able to react with water or hydroxyl ions to generate hydroxyl radicals. Qin et al. [131] showed that the introduction of –NH₂ enhanced the electron density of carbon shell and more electrons were transferred from carbon to the metal oxide, which enhanced the generation of •OH radicals. Nie et al. [134] reported •OH was the main active species in the entire reaction in ultrathin iron–cobalt oxide nanosheets/H₂O₂ system. The main reactive species involved in H-AOPs for TC degradation are shown in Table 3.

Table 3. Synthesis methods for MCs and their leading reactive species during TC degradation.

| Magnetic Materials                  | Synthesis Techniques               | Leading Reactive Species | Removal Rate (%) | Quenchers | Advanced Oxidation Processes | References |
|-------------------------------------|------------------------------------|--------------------------|-----------------|-----------|-------------------------------|------------|
| Fe₃O₄@MSC                           | Co-precipitation process and a calcination process | •OH                      | 99%             | NA        | H-AOPs                        | [135]      |
| Biochar modified CuFeO₂ (CuFeO₂/BC) | Hydrothermal method                | •OH                      | 88%             | NA        | H-AOPs                        | [122]      |
| Fe₃O₄-Cs                            | Co-precipitation                   | •OH                      | 96%             | TBA, KI, BQ and DMPO | H-AOPs | [124] |
| Magnetic core–shell MnFe₂O₄@C        | Hydrothermal synthesis             | •OH                      | 64%             | TBA and BQ | H-AOPs                        | [131]      |
| Fe-MOFs                             | Solvothermal method                | •OH                      | 83%             | NA        | H-AOPs                        | [125]      |
| Magnetic NiFe₂O₄/C                  | Calcination                        | •OH and •O₂⁻            | 97%             | Isopropanol (IPA), 4-hydroxy-TEMPO (TEMPO), and triethanolamine (TEOA) | H-AOPs | [130] |
| yolk-shell nanospheres              |                                    |                          |                 |           |                               |            |
| Fe₃O₄ nanospheres                   | One-pot solvothermal method        | •OH, •O₂⁻, and •HO₂     | 80%             | TBA, KI, BQ | H-AOPs                        | [133]      |
| Magnetic palygorskite nanoparticles (Pal@Fe₂O₃) | Co-precipitation method                   | •OH and •O₂⁻            | 73%             | NA        | H-AOPs                        | [129]      |
| TiO₂/Fe₂O₃                          |                                    |                          |                 |           |                               |            |
| hierarchical porous composites      |                                    |                          |                 |           |                               |            |
| Iron-cobalt oxide nanosheets (CoFe-ONNs) | High-temperature calcination      | •OH and •O₂⁻            | 98%             | TBA        | H-AOPs                        | [136]      |
| FeNi₂/SicO₂/ZnO magnetic nanocomposite | Surfactant-aided co-reduction process | •OH                      | 84%             | TBA        | H-AOPs                        | [134]      |
| MnFe₂O₄@C-NH₂ nanoparticles        | Solvothermal method                | h⁺, •O₂⁻ and •OH        | 100%            | NA        | H-AOPs                        | [137]      |
| Sulfurized oolitic hematite          | Hydrothermal synthesis             | •OH                      | 64%             | TBA and BQ | H-AOPs                        | [131]      |
|                                    | Calcination                        | •OH and •O₂⁻            | 90%             | TBA and p-BQ | H-AOPs                        | [138]      |
Table 3. Cont.

| Magnetic Materials | Synthesis Techniques | Leading Reactive Species | Removal Rate (%) | Quenchers | Advanced Oxidation Processes | References |
|--------------------|----------------------|--------------------------|------------------|-----------|----------------------------|------------|
| Pyrite             | Co-precipitation and high temperature calcination | OH                      | 85%              | NA        | H-AOPs                      | [132]      |
| Mn doped magnetic biochar (MMBC) | Hydrothermal methods | OH                        | 93%              | MeOH (MeOH), TBA and BQ | S-AOPs     | [139]      |
| Magnetic rape straw biochar (MRSB) | Pyrolysis | O$_2^\cdot$ and OH and SO$_4^\cdot$ | 86%              | NA        | S-AOPs                      | [140]      |
| Biochar supported nanosized iron (nFe(0)/BC) | Physical ball milling | OH and SO$_4^\cdot$ | 87%              | NA        | S-AOPs                      | [108]      |
| Fe8BC              | Chemical reduction method | OH and SO$_4^\cdot$ | 98%              | Ethanol (EtOH) and TBA | S-AOPs     | [141]      |
| Nano Fe(0) immobilized mesoporous carbon | One-step method | OH and SO$_4^\cdot$ | 100%             | MeOH and TBA | S-AOPs                      | [142]      |
| Fe-N-BC            | Liquid-phase reduction method | SO$_4^\cdot$ | 92%              | MeOH and TBA | S-AOPs                      | [143]      |
| MS-biochar         | Hydrothermal synthesis | OH                        | 97%              | NA        | S-AOPs                      | [146]      |
| Fe-MOFs            | CuO/MnFe$_2$O$_4$ nanocomposite | OH and SO$_4^\cdot$ | 74%              | MeOH      | S-AOPs                      | [113]      |
| Magnetic C$_3$N$_4$/CuFe$_2$O$_4$/Fe$_2$O$_3$ composite | Co-precipitation | OH and SO$_4^\cdot$ | 91%              | MeOH and TBA | S-AOPs                      | [147]      |
| MnFe$_2$O$_4$ nanoparticles | Sol-gel combustion method | OH and SO$_4^\cdot$ | 89%              | MeOH      | S-AOPs                      | [64]       |
| Magnetic NixFe$_3$-xSO$_4$ Aug-BiFe$_2$O$_3$ MIL-101(Fe)/TiO$_2$ composite | Hydrothermal and calcination method | OH and SO$_4^\cdot$ | 100%             | BQ, EDTA, TBA and IPA | S-AOPs     | [148]      |
| FeO@POCN/CQDs      | Coprecipitation method | OH and SO$_4^\cdot$ | 96%              | EtOH, TBA, p-BQ and | S-AOPs     | [149]      |
| CNx/Fe$_3$O$_4$/SS | Sol-gel method | OH and SO$_4^\cdot$ | 91%              | t-BuOH and MeOH and | S-AOPs     | [150]      |
| Fe$_3$O$_4$ nanocomposites | Solvothermal method | OH                        | 90%              | NA        | S-AOPs                      | [151]      |
| FeO$_2$-NCS-x      | Selfassembly method | OH and SO$_4^\cdot$ | 98%              | Sodium oxalate (SO), BQ and IPA | S-AOPs | [152]      |
| MnFe-LDO-biochar   | Electro-polymerization and Pyrolysis | OH and SO$_4^\cdot$ | 100%             | NA        | S-AOPs                      | [153]      |
| TiO$_2$ decorated on magnetic activated carbon (MAC@Ti) | Hydrothermal method | OH and SO$_4^\cdot$ | 93%              | NA        | S-AOPs                      | [123]      |
| Fe-based metal organic frameworks (MIL-88A) | Solvothermal method | OH                        | 89%              | KI, TBA and sodium azide (NaN3) | S-AOPs | photocatalysis | [123] |
| ZnO/γ-Fe$_2$O$_3$  | Microwave assisted aqueous solution method | OH                        | 86%              | TBA, EtOH, N2 and N2 plus EtOH | S-AOPs | photocatalysis | [156] |
| ZnFe$_2$O$_4$      | Co-precipitation-calcination process | OH and SO$_4^\cdot$ | 98%              | t-BuOH | photocalysis | [155] |
| 3D CoFe$_2$O$_4$/NiFe$_2$O$_4$FeNi$_2$@TiO$_2$ nanocomposite | Hydrothermal method | OH and SO$_4^\cdot$ | 92%              | t-BuOH, EDTA and BQ | S-AOPs | [157]      |

4.2. Sulfate Radical Based Advanced Oxidation Processes (S-AOPs)

S-AOPs generally aroused the interest of researchers since SO$_4^\cdot$ (E$_0$ = 2.6–3.1 V) had a comparable or even higher redox potential than OH (E$_0$ = 1.8–2.7 V) at natural pH [159]. Further, SO$_4^\cdot$ radical own longer half-life than OH, and it can have better mass transfer ability and more stably contact with target contaminant [160]. The MCs can activate persulfate (PS) or peroxymonosulfate (PMS) to produce SO$_4^\cdot$ and OH radicals which well further attacked antibiotics molecules to generate smaller intermediate product. The results showed that SO$_4^\cdot$ played a significant role for TC degradation in the nano Fe$_0$/MC+PS system.
Zhou et al. [161] suggested that some functional structures of biochar (such as pore structure, oxygen-containing groups, and defects) could be beneficial to catalysis. The carboxy and hydroxyl on swine bone derived biochar (BBC) might enhance radical pathway, which can help to generate $\bullet OH$ and SO$_4$'$. Likewise, Huang et al. [139] found that Mn doped magnetic biochar (MMBC) was highly conductive and electron transport existed in TC degradation. The removal efficiency of TC reached 93%, which was much higher than that of the original BC (64%). Huang et al. [140] also observed that magnetic rape straw biochar (MRSB) exhibited 13.2-fold higher reaction rate for activating PS than those of rape straw biochar (RSB) in the MRSB/PS system.

Wan et al. [82] found that Fe-based MOFs could enhance catalyst performance for PS to degrade organic pollutants. He et al. [151] used TiO$_2$-based MOFs composite for TC degradation and the degradation rate was 90.15% in 5 min under xenon lamp irradiation. Lv et al. [162] used hydrothermal and calcination method to prepare g-C$_3$N$_4$@CoFe$_2$O$_4$/Fe$_2$O$_3$ composite. The results showed that MOF-derived CoFe$_2$O$_4$/Fe$_2$O$_3$ could remarkably enhance visible light absorption ability of g-C$_3$N$_4$ and reduce band gap of g-C$_3$N$_4$. Significantly, TC of 99.7%, BPA of 98.1%, SMX of 94.8%, DFC of 97.0%, IBP of 96.1% and OFX of 96.5% could be removed within 80 min.

Li et al. [48] investigated the TC degradation in the Cu/CuFe$_3$O$_4$ activated PS system in which the Cu$^0$ and in-situ generated Cu(I), Cu(II), Cu(III), Fe(II), and Fe(III) ions could activate persulfate to generate SO$_4$' and OH•. Ma et al. [147] used magnetic CuO/MnFe$_2$O$_4$ nanocomposite as a heterogeneous catalyst to activate PS for levofloxacin (LVF) removal. The results revealed that the CuO/MnFe$_2$O$_4$ showed higher catalytic performance than pure CuO, pure MnFe$_2$O$_4$, and other fabricated CuO/MnFe$_2$O$_4$ nanocomposites. Guan et al. [117] suggested that heterogeneous magnetic Ni$_x$Fe$_{3-x}$O$_4$ catalysts could promoted TC degradation in Ni$_x$Fe$_{3-x}$O$_4$/PS system. The results showed that Ni$_{10.6}$Fe$_{2.4}$O$_4$ presented superior catalytic activity performance and catalyze the PS to generate SO$_4$' and HO• to efficiently degrade TC.

In general, SO$_4$' is generally produced by radiolysis, photolysis, pyrolysis, or chemical activation of PMS or PS. The reaction via metal ions and PMS/PS is primarily based on the electron transfer process between the metal ions and the oxidants. Herein, the following mechanisms were found during the TC degradation in MCs/S-AOPs system. Huang et al. [139] found surface oxygen-containing functional groups, and the defect structure of the material and the iron-manganese oxide were active reaction sites of MMBC to activate PS. Wan et al. [82] reported $\bullet O_2$ - and SO$_4$' played an important role in demineralizing the organic pollutants in the Fe-MOFs-D-7.5/PS system. Hu et al. [110] indicated that the transformation rates of Fe(II)/Fe(III) were the main factors to deter the catalytic efficiency of MNPs for PS in the PS/MNPs system. Guan et al. [117] found that high oxidative SO$_4$’ and HO• were the main radical species on TC degradation in the Ni$_{10.4}$Fe$_{2.4}$O$_4$/PS system. Tang et al. [149] suggested that the active radical contribution order could be concluded as follows: SO$_4$’ $\rightarrow$ $\bullet OH$ $\rightarrow$ $\bullet O_2$ - $\rightarrow$ $\cdot$O$_2$. The main reactive species involved during S-AOPs for degradation of TC are shown in Table 3.

4.3. Photocatalysis

Semiconductor can be activated by photon and inspired an electron from the valence band to the conductive band in the photocatalytic system, thus generate electron–hole pairs in the valence band, and the equation could be described as:

$$\text{Semiconductor} + h\nu \rightarrow e^- + h^+$$  \hspace{1cm} (3)

The $e^-$ and $h^+$ could further react with electron donors and acceptors on the surface of semiconductor to generate free radicals [163]. However, there were some limits occurring in the photocatalyst utilization, including low utilization of solar energy [164], high recombination rate of the photogenerated electron–holes [165], low stability, and difficult separation from water [26]. Therefore, the heterogeneous photocatalysts were developed to enhance the efficiency of pollutant degradation.
Azalok et al. [155] prepared a high-efficiency layered double oxide–biochar hybrid (MnFe-LDO–biochar) catalyst to degrade TC in aqueous solution. The characterization results verified that MnFe-LDO–biochar possessed a specific surface area of 524.8 m² g⁻¹, appropriate bandgap (2.85 eV) and a mixture of interconnected pores. The MnFe-LDO–biochar can effectively degrade TC with a removal rate of 98%. Kakavandi et al. [123] found that TiO₂ decorated magnetic activated carbon (MAC@T) coupled with US and UV irradiation could effectively remove TC in aqueous solution. At optimal conditions, over 93% TC was removed in 180 min. Cao et al. [42] synthesized a plural photocatalyst consisting of graphene oxide, magnetite, and cerium-doped titania. The graphene oxide with large specific surface area can adsorb TC onto the catalyst surface, and the radicals activated by cerium-doped titania can rapidly react with adsorbed TC. Furthermore, the intermediates and photocatalytic route of TC degraded by this catalyst was analyzed by liquid chromatography–mass spectroscopy, and three routes were found.

Recently, Fe-based MOFs have also served as photocatalysts [79]. For example, Zhang et al. [156] used a hydrothermal method to synthesize Fe-based MOFs (MIL-88A) as a high-efficiency catalyst for degrading TCH under visible light irradiation. The 200 mL TC with a concentration of 100 mg/L could be entirely degraded and the degradation kinetics fitted well with the pseudo-second-order model. In addition, Wang et al. [165] used Fe₂O₃ to support Bi₂WO₆ at different composite ratio, and it was demonstrated that an appropriate amount of Fe₂O₃ could improve visible-light response and nanospheres morphology of the heterogeneous photocatalyst. Moreover, Semeraro et al. [128] investigated the photocatalytic activity of ZnO/γ-Fe₂O₃ composite catalyst in the photocatalytic degradation of TC. The results clearly showed that the ZnO/γ-Fe₂O₃ composite catalyst presented significant photocatalytic activity with degradation efficiency of 88.52%. Moreover, ZnO was found to play the key role in the photocatalytic process assisted by γ-Fe₂O₃ which enhanced the TC degradation efficiency by 20%.

Nasseh et al. [137] prepared magnetically separable FeNi₃/SiO₂/ZnO nano-composite to degrade TC under simulated sunlight. Khodadadi et al. [53] prepared FeNi₃@SiO₂@TiO₂ nanocomposite to remove TC by photo-catalytic degradation in simulated wastewater. The results showed that ZnFe₂O₄ catalyst had both microwave–catalytic and visible-light photocatalytic activities and 91.6% of TCH degradation was obtained in the MW/MEDL/ZnFe₂O₄ system in 4 min.

Among many photocatalytic materials, g-C₃N₄, as a typical metal-free organic semiconductor photocatalyst, has attracted much attention in the field of photocatalysis due to convenient synthesis, nontoxicity, low-cost, and suitable band gap. Sun et al. [34] found that the N-deficient g-C₃N₄ (CNₓ)/PS system displayed a high efficiency in the photocatalytic process of TC degradation. Wang et al. [166] reported the C₃N₄@MnFe₂O₄-G composites showed a superior catalytic activity with 94.5% removal of metronidazole that was almost 3.5 times as high as that of the pure g-C₃N₄ which could be attributed to the synergistic promoting effect of the favorable adsorption.

Overall, MCs were efficient photocatalysts and could effectively remove TC in aqueous solution under photocatalytic process. The reactive oxygen species such as •O₂⁻, •OH, and H₂O₂ played important role to degrade TC, because the reactive oxygen species attacked TC molecules and converted it into less-toxic intermediates or completely degraded into the CO₂ and H₂O [57]. The following mechanisms existed in the degradation of TC by MCs during the photocatalytic process. Azalok et al. [155] found that the TC photodegradation mechanism was induced mainly by •OH and SO₄²⁻ while h⁺ and •O₂⁻ contributed partly to the TC decomposition in the MnFe-LDO–biochar system. Kakavandi et al. [123] observed that •OH and ¹O₂ were main oxygen species in (MAC@T) coupling with the US/UV irradiation system. In addition, Pang et al. [157] suggested that h⁺ was the main active species for TCH degradation and little •O₂⁻ active species generated in MW/MEDL/ZnFe₂O₄ system. Wang et al. [166] reported that the h⁺, •O₂⁻, SO₄²⁻, and •OH were responsible for the TC decomposition in C₃N₄@MnFe₂O₄-G system.
The main reactive species involved during the photocatalytic removal of TC on different reaction systems are shown in Table 3.

AOPs could efficiently degrade tetracycline through active free radicals attacking the chain structure of TC. In this section, the application of MCs on the removal of TC via AOPs is fully presented. $\bullet$O$_2^{-}$, $\bullet$OH, and SO$_4^{\bullet-}$ played important roles to degrade TC during AOPs. Moreover, the MCs could degrade TC through Fenton, Fenton-like, photo-Fenton, photocatalysis processes, and sulfate-based AOPs, and their performances were superior to the homogenous catalyst due to the high stability and synergistic effect between components. For H-AOPs, $\bullet$OH was the most important active species and $\bullet$OH production rate determines the degradation efficiency of TC in the system. For S-AOPs, the system had strong stability and tetracycline degradation efficiency, because the system can produce a variety of active species. For photocatalysis, the organic–inorganic composite magnetic photocatalyst has a smaller band gap, and thus has a stronger catalytic ability, because it improves the utilization rate of the light source.

5. Synergistic Effects between MC Components for Degrading TC

In many cases, the combination of two components could create synergistic effect to improve the degradation efficiency of TC because certain materials can prevent magnets from agglomeration. Lai et al. [120] fabricated a MnFe$_2$O$_4$/biochar composite, in which the biochar was fictionalized to prevent magnets from the aggregation proven by SEM. Likewise, Pi et al. [167] synthesized Fe$_3$O$_4$ magnetized biochar, and the SEM images showed that the Fe$_3$O$_4$ was uniformly coated on the biochar surface. Li et al. [124] observed that chitosan biopolymer serving as support can prevent the agglomeration of Fe$_3$O$_4$ NPs and the H$_2$O$_2$ could be activated by the strong synergistic effect between Fe-based groups and carbon matrix, thereby increasing the degradation efficiency of TC. Huang et al. [139] found that MRSB could greatly accelerate generation of SO$_4^{\bullet-}$, $\bullet$OH, and $\bullet$O$_2^{-}$ to enhance the TC degradation efficiency. Moreover, Lv et al. [162] reported the introduction of MOFs-derived CoFe$_2$O$_4$/Fe$_2$O$_3$ provided a new approach for generating radical species, and the k value of g-C$_3$N$_4$@CoFe$_2$O$_4$/Fe$_2$O$_3$ was the highest (0.0524 min$^{-1}$), which was nearly 26.2 times as high as that of g-C$_3$N$_4$. These results fully indicated that MOFs-derived CoFe$_2$O$_4$/Fe$_2$O$_3$ could greatly enhance photocatalytic efficiency of g-C$_3$N$_4$. Furthermore, the surface interaction of components can accelerate the separation of photogenerated electron–hole pairs, thus enhancing catalytic activity. For instance, Zhu et al. [168] synthesized a nanoreactor (MS@FCN) whose core was Fe$_3$O$_4$ magnetized graphitic carbon nitride (Fe$_3$O$_4$g-C$_3$N$_4$) and a shell of mesoporous silica. The prominent electrical conductivity of Fe$_3$O$_4$ restrained the recombination of electron–hole in the graphitic carbon nitride, which bifitted the charge separation and lengthened the life of the photocarrier, thus facilitating the generation of radicals, while the mesoporous silica shell provided a big surface area and refractive condition that enhanced the photocatalytic activity. In addition, He et al. [151] observed that TiO$_2$ introduced in the composite played an important role in the degradation process, in which TiO$_2$ had a synergistic effect with Fe$_3$ to generate Fe$_3^{2+}$, Ti$^{2+}$ and radicals Fe$_2^{2+}$ reacted with PS to produce Fe$_3^{3+}$ and a number of $\bullet$OH to degrade TC in TiO$_2$-based metal–organic frameworks system. Pu et al. [146] found surface-bound Fe(II) acted as the main active site to provide electrons for PS or dissolved oxygen and effectively activate PS.

To sum up, the magnetic heterogeneous catalysts could bring the following advantages for the degradation of TC via AOPs: They (1) facilitated the separation and enhanced the stability of catalysts; (2) promoted the transformation of photogenerated charge carriers, depressed the recombination of electron–hole pairs, and prolonged the lifetime of the photogenerated holes, thus improved the catalytic ability; (3) prevented the aggregation of catalysts; (4) increased the production of free radicals; and (5) promoted the generation of free radicals as an intermediate.
6. Reusability of MCs

For a catalyst, recyclability is an important characteristic, which relates to its cost of actual operation [169]. It is hard to restrain the decline in degradation performance because of the loss of quality, but thanks to the magnetic separation method, which lost less mass compared to filtration, sediment, and centrifugation [42], the magnetic catalysts showed satisfactory recyclability. Zhong et al. [43] synthesized a ZnFe$_2$O$_4$ magnetized Bi$_2$WO$_6$ to degrade TCH and observed that this material could still reach 81.52% removal efficiency after five cycles. Likewise, Guan et al. [117] tested the reusability of obtained magnetic Ni$_{0.6}$Fe$_{2.4}$O$_4$ catalyst and discovered only a 3% decrease in the degradation rate after four times of reuse. Furthermore, the X-ray diffraction patterns of magnetic Ni$_{0.6}$Fe$_{2.4}$O$_4$ catalyst before and after four times of reuse indicated that this material had high stability. Yu et al. [135] found that the UV assisted heterogeneous Fenton-like process in Fe$_3$O$_4$@MSC improved the cycle of Fe$^{3+}$/Fe$^{2+}$ and activated the interfacial catalytic site, which eventually realized the enhancement of degradation and mineralization to TC. Wang et al. [138] reported that sulfured oolitic hematite (SOH-600) exhibited an excellent recycling performance and a high catalytic efficiency (>90%) after five cycles. Ren et al. [158] suggested that the degradation efficiency of TC was detected to be as high as 85% only within 10s and the degradation rate can remain above 90% after five cycles under the presence of 3D CoFe$_2$O$_4$/N-rGA and PMS.

7. Recommendations

Attention on applying MCs on TC removal in aqueous solutions has increased yearly due to the convenient separation of MCs from aqueous solution, and the stable, highly efficient removal performance. Although the MCs have been proven to be excellent heterogeneous catalysts, some research gaps still exist in the present research. Based on the literature, the following key perspectives should be addressed in the future: (1) The most common magnets used to synthesize MCs was Fe$_3$O$_4$, while other magnets, such as CoFe$_2$O$_4$, NiFe$_2$O$_4$, and MgFe$_2$O$_4$ with the better performance have not well been developed and utilized; (2) Less previous studies mentioned the cost of materials, but cost is a major obstacle to commercial applications; the cost analysis of materials should be better considered; (3) Further investigations should be carried out to attain the maximum possible efficiency and effectiveness regarding synthesis, application, and recycling of MCs; (4) The preparation of MCs is complicated and more attention should be paid to the green synthesis methodologies, e.g., ball milling method and microwave-assisted heating; (5) The pH of the reaction system has a great influence on the removal of TC from MCs. It is necessary to strengthen the adaptation range of the MCs to the pH of the system and improve the practical application ability of MCs; (6) Bi-pollutant, tri-pollutant, or multipollutant systems based on TC pollution system were more complicated and need more investigation; (7) MCs have a certain mass loss in the recycling process, and it is very necessary to develop novel recycling technologies; and (8) At present, the application of MCs was in the laboratory scale, and the governance of MCs in piratical engineering use requires more attention.

8. Conclusions

Previous studies have demonstrated that MCs have exceptional performance in removing TC. To date, carbon-based MCs, polymer-based MCs, and MOFs have been mainly used to remove TC in aqueous solution. Moreover, magnetic graphene and metal-modified magnetic biochar showed excellent performance in removing TC from aqueous solutions. The adsorption of TC on graphene-based MCs mainly accorded with the Langmuir model, while the adsorption of TC on biochar-based materials mainly conformed to the Freundlich model. In addition, Polymer-based MCs were generally suitable for the pseudo-second-order kinetic model, which mainly accorded with Langmuir model. Complexation, cation exchange, electrostatic attraction, hydrogen bonding, and π-π interaction played important roles to adsorb TC.
AOPs are considered as an efficient, rapid, and environmentally-friendly approach for TC degradation. •O$_2^-$, •OH, and SO$_4^{•-}$ were the main free radicals to degrade TC during AOPs. For H-AOPs, MnFe$_2$O$_4$/biochar composite, Fe-MOFs, and Fe$_3$O$_4$-based materials had excellent catalytic ability in the photo-Fenton system. For S-AOPs, G-C$_3$N$_4$/CoFe$_2$O$_4$/Fe$_2$O$_3$ composite, Fe-MOFs, and Ni$_x$Fe$_{3-x}$O$_4$ catalysts had strong catalytic ability and the active radical contribution order could be concluded as follows: SO$_4^{•-}$ > •OH > •O$_2^-$ > 1O$_2$ in PS system. For photocatalysis, the g-C$_3$N$_4$(CN$_x$)/PS system had a strong catalytic ability for TC degradation.

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