REVIEW ARTICLE

A review of Pharmaceuticals removal from water resources using magnetic iron-based nanomaterials

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Abstract: The presence of pharmaceuticals in water resources is a growing concern worldwide due to their potential health impacts on aquatic life and humans. Therefore, there is a need to develop effective and sustainable technologies for removing these contaminants from water and wastewater. Magnetic nanomaterials have emerged as promising materials for this purpose due to their fast kinetics, easy magnetic separation, and reuse. This review is important as it highlights the significance of developing sustainable technologies using magnetic iron-based nanomaterials for removing pharmaceutical contaminants from water resources. This review investigated the application of magnetic nanomaterials for removing pharmaceuticals from water resources through adsorption and advanced oxidation processes. Here, the synthesis and characterization of magnetic nanomaterials and analytical detection techniques were evaluated. The review findings indicate that magnetic nanomaterials effectively removed pharmaceuticals from water through adsorption and advanced oxidation processes. More importantly, the removal processes remained effective for many cycles. However, only 22% of the studies demonstrated the application of magnetic nanomaterials on real water samples, as 78% stopped at experiments using distilled water in the laboratory. Further research on multi-component systems and real water samples is necessary to fully evaluate the potential of magnetic nanomaterials for pharmaceutical removal from water resources.

Keywords: Contaminants of emerging concern; Pharmaceuticals; Magnetic Iron based nanomaterials; Adsorption; Advanced oxidation process.

1. INTRODUCTION

Globally, pharmaceuticals have been frequently detected in water resources such as surface water, groundwater and industrial wastewater in the last decade. In Africa, different classes of pharmaceuticals, such as antibiotics, analgesics, anticonvulsant, anti-inflammatory, and antiviral drugs etc., were detected in surface water, groundwater and industrial wastewater from Uganda (Dalahmeh *et al.*, 2020; Nantaba *et al.*, 2020), Kenya (K'oreje *et al.*, 2016; K'oreje *et al.*, 2012,), Nigeria (Folarin *et al.*, 2019; Jennifer *et al.*, 2020) and South Africa (Gumbi *et al.*, 2016; Hlengwa & Mahlambi, 2020; Madikizela *et al.*, 2017; Sibeko *et al.*, 2019). More details on occurrences, distributions and risk assessments of pharmaceuticals in water resources across five African regions are well documented by (Shehu *et al.*, 2022). Coincidentally, similar trends of pharmaceuticals detected in African water resources were also found in Greece (Kosma *et al.*, 2014; Papageorgiou

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et al., 2016), Czech republic (Buriánková *et al.*, 2021; Golovko *et al.*, 2014), Poland (Kapelewska *et al.*, 2018; Migowska *et al.*, 2012; Styszko *et al.*, 2021), Canada (Guerra *et al.*, 2014), India (Diwan *et al.*, 2009; Mohapatra *et al.*, 2016), Iran (Mirzaei *et al.*, 2018), Pakistan (Ashfaq *et al.*, 2016), Qatar (Al-maadheed *et al.*, 2019), Singapore (Tran *et al.*, 2016), Spain (Gros *et al.*, 2013), Croatia (Senta and Ahel 2012), Belgium (Lorenzo *et al.*, 2018), Italy (Verlicchi *et al.*, 2012, 2014), Germany (Rossmann *et al.*, 2018), Malaysia (Omar *et al.*, 2019), USA (Karthikeyan & Meyer, 2006), Vietnam (Nguyen *et al.*, 2015), China (Hanna *et al.*, 2018), Japan (Murata *et al.*, 2011), Iraq (Al-Khazrajy & Boxall, 2016) and Sweden (Baresel *et al.*, (2015). Enough evidence to declare pharmaceutical pollution as a global issue affecting water resources is documented by (Omuferen *et al.*, 2022).

Due to the regular occurrence of pharmaceuticals in global water resources, its risks, such as the development of antibiotic resistance and toxicity to aquatic life, it is imperative to develop appropriate technologies that can specifically removal pharmaceuticals from water (Shehu *et al.*, 2022; Wang & Wang, 2016, 2019, 2022). The conventional water and wastewater treatment methods are advanced oxidation, reverse osmosis, ultrafiltration, electrochemical methods, coagulation-flocculation, precipitation, ion exchange, and adsorption(Ajala *et al.*, 2018). However, these technologies are not effective in the removal of pharmaceuticals from water and wastewater, and a number of these compounds are frequently detected in water bodies despite the presence of water treatment facilities (Wang *et al.*, 2019; Wang & Chu, 2016; Wang & Zhuan, 2020). One promising approach in water or wastewater treatment is the use of nanotechnology for contaminant removal.

Nanotechnology involves the design, synthesis, characterization, and applications of structures, devices and systems at the nanometer scale (Porwal & Sharma, 2016; Qu *et al.*, 2013). Nanomaterials have attracted attention due to unique properties such as the high surface area to volume ratio, surface reactivity, and ability to be functionalized for a specific target. Thus, nanomaterials are used as nanoadsorbents or nanocatalysts for the removal of a wide range of pollutants, including pharmaceuticals, from water and wastewater (Bhattacharya *et al.*, 2013; Chakraborty *et al.*, 2022; Kar *et al.*, 2021, 2022; Kaushik *et al.*, 2021; Lamayi *et al.*, 2018; B. Sharma *et al.*, 2021; Shehu & Lamayi, 2019; N. Singh *et al.*, 2022). In recent times, numerous reported applications of nanoparticles for the removal of various pharmaceuticals in water through adsorption processes (Choina *et al.*, 2014; Dada *et al.*, 2021; Rosli *et al.*, 2021; Xing *et al.*, 2020) and advanced oxidation processes (Hojamberdiev *et al.*, 2020; Zeng *et al.*, 2018; Zhu *et al.*, 2019). However, the significant setbacks of nanotechnology are the separation of the nanomaterials after treatment, instability and possible toxicity that could arise due to the nanoparticles' choice (Bhattacharya *et al.*, 2013). These problems can be solved by using magnetic nanomaterials, the development of nanocomposites and choosing less or non-toxic nanomaterials.

Magnetic Iron based nanomaterials include; nano zerovalent Iron (Li *et al.*, 2017; Rosická & Šembera, 2011; Vecchia *et al.*, 2009), magnetite (Fe_3O_4) (Saeed *et al.*, 2020), maghemite (Fe_2O_3) (Aragaw and Aragaw 2021) and magnetic metal ferrites (Bao *et al.*, 2013). Magnetic iron nanoparticles are widely used in environmental remediation because it is less or non-toxic and sustainable. Sustainability is due to magnetism that provides easy recovery and reuse. Moreover, magnetic iron nanoparticles possess fast kinetics and high adsorptive and catalytic properties. Noteworthy, the magnetic saturation requirement of 1.0 emu/g is needed for any magnetic materials expecting magnetic separation after water treatment (Li *et al.*, 2019).

Therefore, this study aimed to review recent contributions of nanotechnology with the aid of Magnetic iron nanoparticles toward treating water-containing pharmaceuticals. Furthermore, various synthesis methods and characterization techniques that are commonly used for Magnetic iron nanoparticles were reviewed. More importantly, this review is limited to applications of Magnetic iron nanoparticles in the advanced oxidation process (AOP) and adsorption process of conventional water treatment.

2. METHOD

The articles were gathered from search engines and database such as Scopus, web of science, google, pumped, Medline and google scholar from 2009 to 2021, Fig.1. The keywords that were used includes pharmaceutical emerging pollutant/contaminants, magnetic nanoparticle/nanomaterials, adsorption, advanced oxidation processes, and water/wastewater. From the literature search, 1000 publications were found. However, 928 articles were excluded

based on; 1) Articles that exclusively investigate the application of magnetic nanomaterials in the removal of emerging pollutants other than pharmaceutical pollutants, 2) Articles that exclusively investigate the application of magnetic nanomaterials in the removal of heavy metals, dyes, microorganism and nutrients other than pharmaceutical emerging pollutants. Hence, only 72 articles (Fig.1) were studied in this review based on the application of magnetic nanomaterials in the removal of pharmaceuticals from water through adsorption and advanced oxidation processes.

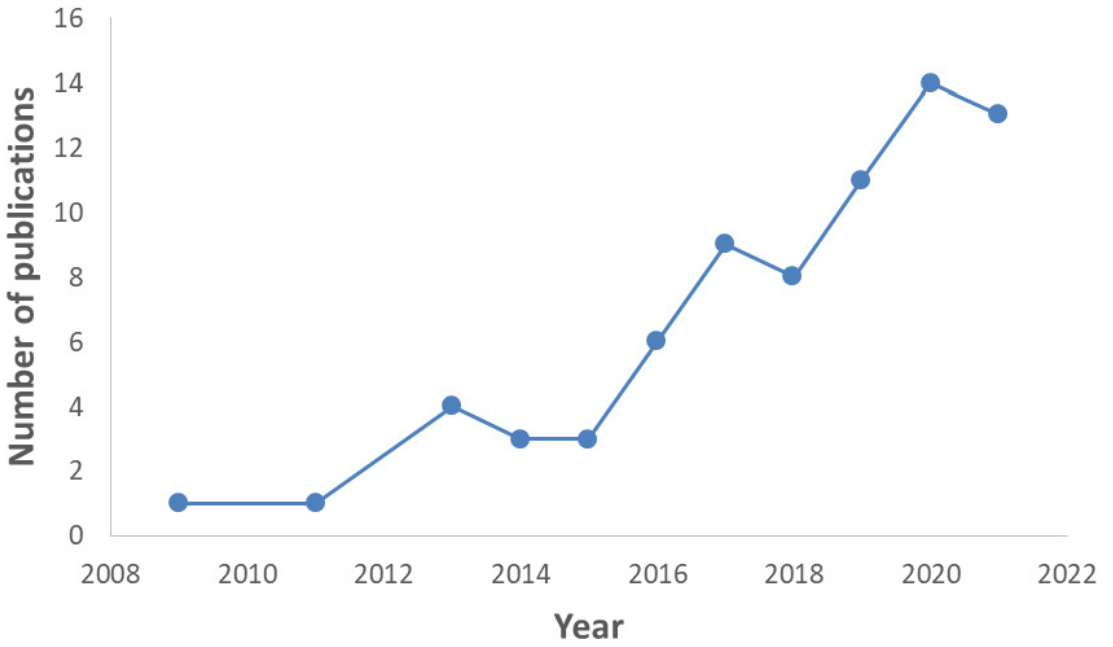

Figure 1. Yearly distribution of publications.

3. SYNTHESIS OF MAGNETIC IRON-BASED NANOMATERIALS

An essential component of nanoscience and nanotechnology is synthesizing materials like magnetic nanoparticles (Balakrishnan *et al.*, 2021; Bao *et al.*, 2014; Lima *et al.*, 2014; Liyanage *et al.*, 2020; Niu *et al.*, 2011; Vicente-Martínez *et al.*, 2020). Only when nanostructured materials are made available with appropriate size, shape, morphology, crystal structure, and chemical composition are new physical attributes and uses of nanoparticles possible (Balakrishnan *et al.*, 2021; Bao *et al.*, 2014; Lima *et al.*, 2014; Liyanage *et al.*, 2020; Niu *et al.*, 2011; Vicente-Martínez *et al.*, 2020). The qualities of a material determine how well it performs. The atomic structure, composition, microstructure, flaws, and interfaces are all influenced by the thermodynamics and kinetics of the synthesis, and these factors, in turn, depend on the characteristics. Many publications have reported various syntheses methods including co-precipitation (47%), hydrothermal (14%), sol-gel (13%), impregnation (9%), microwave-assisted (6%), green (3%), precipitation (3%), thermal decomposition (1%), sonochemical (1%), solvothermal (1%), chemical deposition (1%) and combustion (1%), Fig. 2. Magnetic iron-based nanomaterials synthesized through these methods have shown promising results in various water and wastewater treatment applications due to their unique properties such as high surface area, magnetic properties, and stability in harsh environments. Among the methods, co-precipitation, hydrothermal and sol-gel are known to be simple and promote surface modification of the nanomaterials. Hence, the methods were widely used in synthesizing nanomaterials for water and wastewater applications. However, co-precipitation is mostly used in nanomaterials syntheses because it can occur at ambient temperature.

However, a brief description of each synthesis is given below. Co-precipitation is a widely used method for synthesizing magnetic iron-based nanomaterials (Foroughi *et al.*, 2019; Huang *et al.*, 2020; Liyanage *et al.*, 2020; Malakootian *et al.*, 2018; Malakootian & Shiri, 2021; Wang, *et al.*, 2020; Yegane Badi *et al.*, 2018). This method involves the

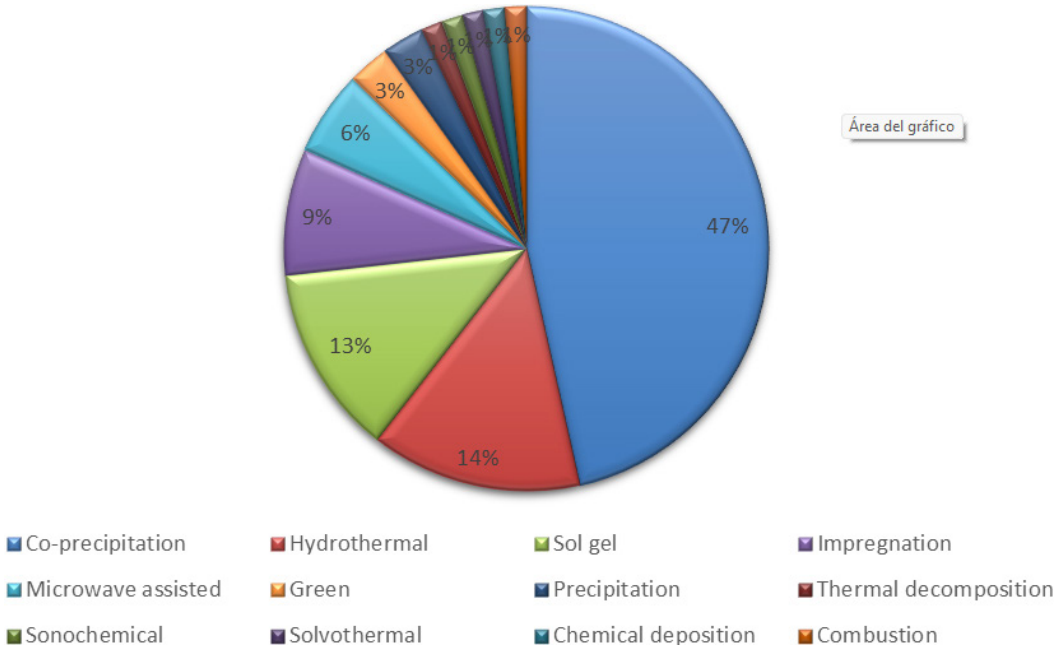

Figure 2. Synthesis of magnetic nanomaterials by different methods.

simultaneous precipitation of iron salts and a base to form iron oxide nanoparticles. The process can be performed at room temperature or higher temperatures, and the particle size can be controlled by adjusting the reaction conditions (Attia *et al.*, 2013; Cusioli *et al.*, 2020; D'Cruz *et al.*, 2020; Ghosh *et al.*, 2013; Kakavandi *et al.*, 2014; Mashile *et al.*, 2020; Mohammadi *et al.*, 2021; Olusegun & Mohallem, 2020; Rocha *et al.*, 2021; Zhang *et al.*, 2017). Impregnation involves the deposition of iron oxide nanoparticles onto a support material by impregnating the support material with a solution containing iron salts. The process is typically performed at room temperature, and the particle size can be controlled by adjusting the reaction conditions(Liu *et al.*, 2019; Mahmoud *et al.*, 2021; Nodeh *et al.*, 2018; Vicente-Martínez *et al.*, 2020). Sol-gel synthesis involves the formation of a colloidal suspension of iron oxide nanoparticles by hydrolysis and condensation of metal alkoxides. The process is typically performed at low temperatures, and the particle size can be controlled by adjusting the reaction conditions (Amraei *et al.*, 2016; Duan *et al.*, 2019; Hayasi & Saadatjoo, 2017; Kollarahithlu & Balakrishnan, 2019; Kumar, Khan, *et al.*, 2018; Mostafaloo *et al.*, 2020; Parashar *et al.*, 2019; Peralta *et al.*, 2021; S. F. Soares *et al.*, 2019). Hydrothermal synthesis involves the use of high-pressure and high-temperature conditions to promote the growth of iron oxide nanoparticles. The process is typically performed in a sealed vessel, and the particle size can be controlled by adjusting the reaction time, temperature, and pressure (Bao *et al.*, 2013; Lu *et al.*, 2016; Malakootian *et al.*, 2019; Mao *et al.*, 2016; Soares *et al.*, 2019). Microwave-assisted synthesis involves the use of microwave radiation to promote the growth of iron oxide nanoparticles. The process is typically performed at room temperature, and the particle size can be controlled by adjusting the reaction time and power (Nasiri, Tamaddon, Hossein, *et al.*, 2019; Nasiri, *et al.*, 2019; Tamaddon *et al.*, 2020). Green synthesis involves the use of natural plant extracts or other environmentally friendly materials to synthesize iron oxide nanoparticles. The process is typically performed at low temperatures, and the particle size can be controlled by adjusting the reaction conditions (Stan *et al.*, 2017; Ye *et al.*, 2021).

Precipitation involves the precipitation of iron salts from a solution to form iron oxide nanoparticles. The process is typically performed at room temperature, and the particle size can be controlled by adjusting the reaction conditions (Dehghan *et al.*, 2018; Oliveira *et al.*, 2017). Thermal decomposition involves the decomposition of iron-containing precursors at high temperatures to form iron oxide nanoparticles. The process is typically performed at high temperatures, and the particle size can be controlled by adjusting the reaction conditions (Li, Ng, *et al.*, 2017). Sonochemical synthesis involves the use of ultrasound waves to promote the growth

of iron oxide nanoparticles. The process is typically performed at room temperature, and the particle size can be controlled by adjusting the reaction time and power (Bao *et al.*, 2014). Solvothermal synthesis involves the use of a solvent at high temperatures and pressures to promote the growth of iron oxide nanoparticles. The process is typically performed in a sealed vessel, and the particle size can be controlled by adjusting the reaction time, temperature, pressure, and solvent type (Silva *et al.*, 2021). Chemical deposition involves the deposition of iron oxide nanoparticles onto a substrate using a chemical reaction. This method typically involves the use of a reducing agent to reduce the iron ions to form iron oxide nanoparticles on the substrate surface. Chemical deposition is a relatively simple and low-cost method for synthesizing magnetic iron-based nanomaterials. However, the particle size and distribution can be difficult to control using this method (Sayadi & Ahmadpour, 2021). Combustion synthesis involves the use of a fuel and an oxidizer to rapidly heat and combust a mixture of metal salts to form metal oxide nanoparticles. This method is typically performed at high temperatures and produces highly crystalline nanoparticles with a narrow size distribution (Al-Anazi *et al.*, 2020b). Combustion synthesis is a relatively simple and low-cost method for synthesizing magnetic iron-based nanomaterials. However, the high-temperature conditions can lead to the formation of impurities and defects in the nanoparticles.

4. CHARACTERIZATION OF MAGNETIC IRON-BASED NANOMATERIALS

Characterization provides comprehensive information on nanomaterials' particle size, shape, composition and magnetic properties. Transmission electron microscopy (TEM), scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), Brunauer-Emmett-Teller (BET) surface area analyzer, Vibrating sample magnetometer, and powder X-ray diffraction (XRD) techniques are a few of the methods frequently used in the literature for magnetic nanoparticle characterization. FTIR spectra reveal the chemical linkages between the magnetic core and the organic surface layer (Bao *et al.*, 2014; Malakootian *et al.*, 2019; Nasiri *et al.*, 2019; Soares *et al.*, 2019; Tamaddon *et al.*, 2020; Wang *et al.*, 2020). Scanning electron microscope/Energy dispersive X-ray spectroscopy (SEM/EDX) can be used to analyze magnetic particles' surface covering and determine their constituent elements (Balakrishnan *et al.*, 2021; Liyanage *et al.*, 2020; Malakootian *et al.*, 2018; Noroozi *et al.*, 2020; Solís *et al.*, 2021; Yegane *et al.*, 2018). Powder XRD is used to characterize the bulk magnetic nanoparticles and identify their crystal structure (Chen *et al.*, 2018; Malakootian and Shiri 2021; Parashar *et al.*, 2019; Sayadi and Ahmadpour 2021). TEM provides candid pictures that can be used to determine the size and shape of nanoparticles (Akkari *et al.*, 2018; Jonidi *et al.*, 2017; Kumar *et al.*, 2018; Mahmoud *et al.*, 2021; Vicente-Martínez *et al.*, 2020). A vibrating sample magnetometer is used to measure the magnetic properties of magnetic materials (Kumar *et al.*, 2018; Lai *et al.*, 2019; Mao *et al.*, 2016; Nodeh *et al.*, 2018). Dynamic light scattering (DLS) is used to investigate the surface charge of magnetic nanomaterials in solution (Balakrishnan *et al.*, 2021; Lima *et al.*, 2014; Malakootian & Shiri, 2021; Olusegun *et al.*, 2021; Parashar *et al.*, 2019; Sayadi & Ahmadpour, 2021). Thermogravimetric analysis (TGA) is used to measure the thermal properties of magnetic nanomaterials (Nawaz *et al.*, 2019; Vicente-Martínez *et al.*, 2020; Zhang *et al.*, 2017). Brunauer-Emmett-Teller (BET) surface area analysis is the multi-point measurement of an analyte's specific surface area (m2/g) using gas adsorption analysis, in which a solid sample is either suspended in a predetermined gaseous volume or continually flowing over by an inert gas such as nitrogen (Kakavandi *et al.*, 2016; Kollarahithlu and Balakrishnan 2019; Li *et al.*, 2017). Energy band gap of magnetic nanomaterials are characterized by DRS/UV/VIS spectrophotometer (Kumar *et al.*, 2018; Malakootian *et al.*, 2019; Nasiri *et al.*, 2019; Nawaz *et al.*, 2019; Sayadi and Ahmadpour 2021). In this review, the percentage of characterization techniques reported in different studies are presented in Fig. 3. However, since each characterization technique present a unique and independent property, the reason for high and low percentage utilization could be due to availability of the instrument to the researcher's disposal. Noteworthy, TGA and DRS/UV/VIS spectrophotometer characterization techniques were observed in unique ways. TGA techniques were widely in characterizing activated carbon and organic molecules modified magnetic nanomaterials. Whereas, DRS/UV/VIS spectrophotometer was used in determining energy band gap for magnetic nanomaterials used in advanced oxidation processes (AOPs).

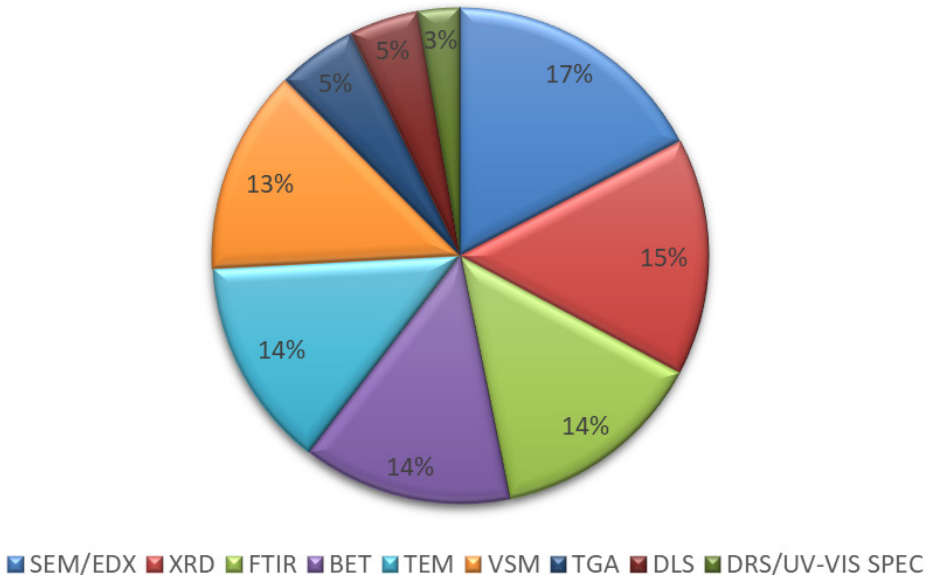

Figure 3. Characterization techniques.

5. ANALYTICAL DETECTION TECHNIQUES

Analytical techniques such as gas and liquid chromatography coupled with UV or mass detectors are used for the identification and quantification of residual concentrations of pharmaceutical emerging pollutants. Also, UV/Visible spectrophotometer is used for the quantification of pharmaceuticals. For adsorption studies, 23, 15 and 1 publications reported quantification of pharmaceutical emerging pollutants using UV/Visible spectrophotometer, HPLC/UV/MS and GC//MS respectively, Fig. 4. Similarly, for advanced oxidation processes (AOPs), 7, and 26 publications reported quantification of pharmaceutical emerging pollutants using UV/Visible spectrophotometer, and HPLC/UV/MS respectively, Fig. 5. In adsorption studies, UV/Visible spectrophotometer was the most frequent used technique because the target is only quantifications while in AOPs, HPLC/UV/MS was frequently used because the goals were quantifications and identifications of degradation products.

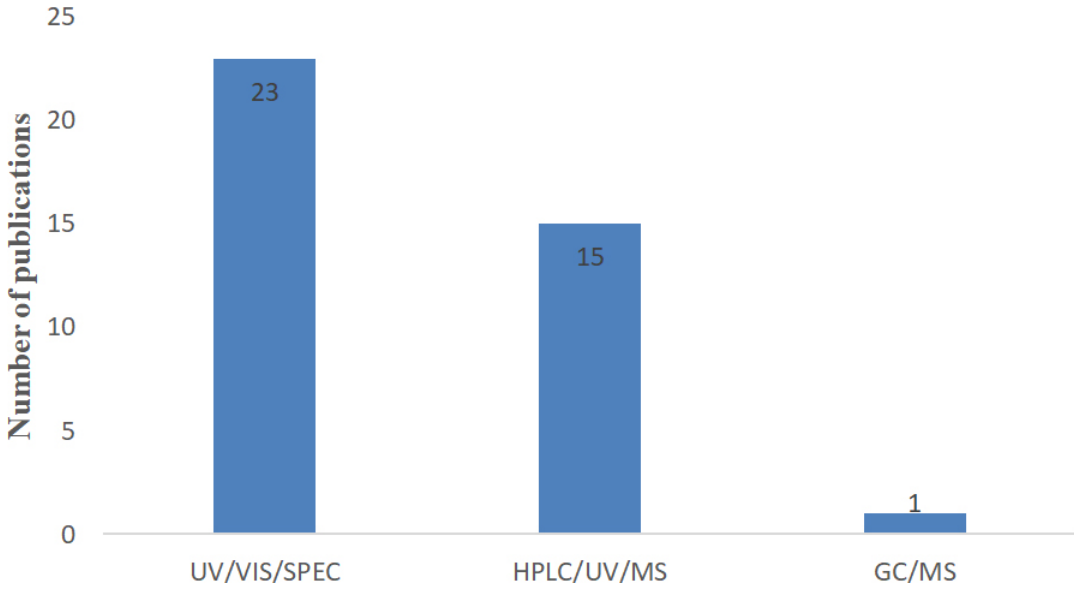

Figure 4. Analytical detection techniques for adsorption studies.

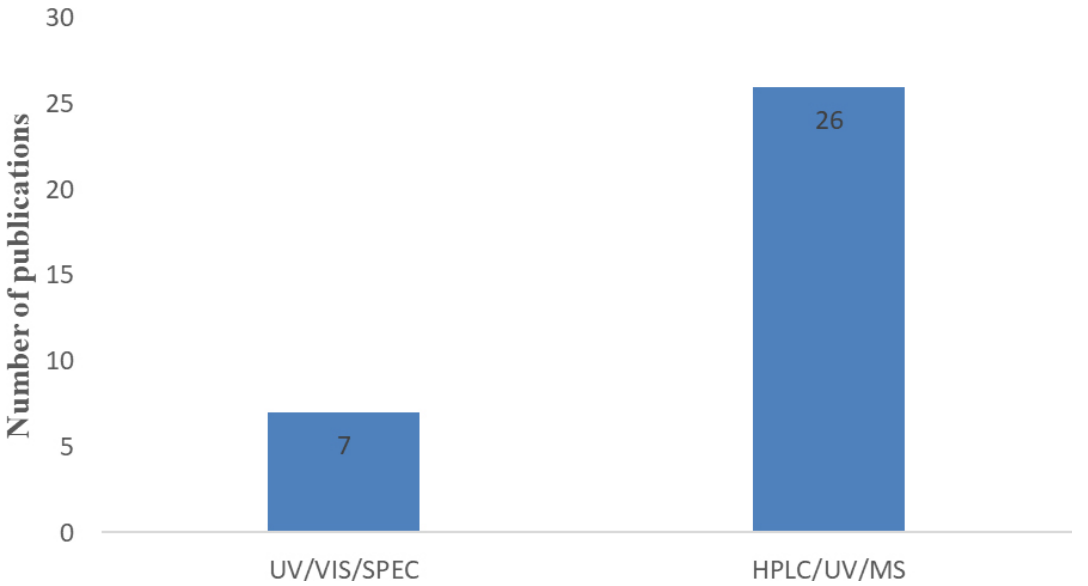

Figure 5. Analytical detection techniques for advanced oxidation processes.

6. STABILITY AND COLLOIDAL STABILITY OF IRON-BASED MAGNETIC NANOMATERIALS FOR WATER AND WASTEWATER TREATMENT

Iron-based magnetic nanomaterials have shown great water and wastewater treatment potential due to their unique magnetic properties and high reactivity(Aragaw *et al.*, 2021). However, these materials' stability and colloidal stability are critical factors that need to be considered for their efficient application. In terms of stability, iron-based magnetic nanomaterials are generally stable in aqueous solutions. However, iron-based magnetic nanomaterials are exposed to various environmental conditions in water and wastewater treatment, such as pH, temperature, and ionic strength, which can affect their stability (Mylon *et al.*, 2004). For example, at low pH values, iron-based magnetic nanomaterials can undergo dissolution, which can lead to a decrease in their effectiveness for water treatment. Colloidal stability is another important factor influencing iron-based magnetic nanomaterials' performance in water and wastewater treatment (Marinin, 2012). The colloidal stability of these materials refers to their ability to remain suspended in solution without aggregation or settling (Wu *et al.*, 2008). Colloidal stability can be affected by factors such as surface charge, particle size, and the presence of other ions (Mylon *et al.*, 2004). The colloidal stability of these materials can be improved by modifying their surface properties through functionalization with organic or inorganic compounds (Akawa *et al.*, 2020, 2021). Surface modification can enhance the electrostatic and steric repulsion between particles, preventing them from agglomerating and improving their colloidal stability (Mao *et al.*, 2016).

The stability of these materials can be evaluated by measuring their aggregation behavior under different conditions. For example, dynamic light scattering (DLS) and zeta potential measurements can be used to determine these materials' size distribution and surface charge (Balakrishnan *et al.*, 2021; Lima *et al.*, 2014; Malakootian & Shiri, 2021; Olusegun *et al.*, 2021; Parashar *et al.*, 2019; Sayadi & Ahmadpour, 2021).

7. PHARMACEUTICALS REMOVAL FROM WATER RESOURCES

In recent time, the demand for pharmaceuticals for humans and animals have increased due to the rapid increase of the global population. Conversely, this leads to an increase in pharmaceutical production. The endpoint of pharmaceutical residue from pharmaceutical industrial wastewater and solid waste is water resources. These have tremendous negative effects on aquatic and terrestrial lives. To regenerate water resources, there is a need to look for the best and most sustainable technology that can remove pharmaceuticals from various sources such as freshwater, groundwater, seawater, and wastewater, etc. Therefore, magnetic nanomaterials stand as the best materials due to their effectiveness

in adsorption and degradation of pharmaceuticals, easy separation from treated water and reuse for several cycles with losing their capacity. The schematic representation of magnetic nanomaterials in the removal of pharmaceuticals from water through adsorption process and advanced oxidation processes is given in Fig. 6. More details of the adsorption process and advanced oxidation processes for the removal of pharmaceuticals are given in sections 7.1 and 7.2, respectively.

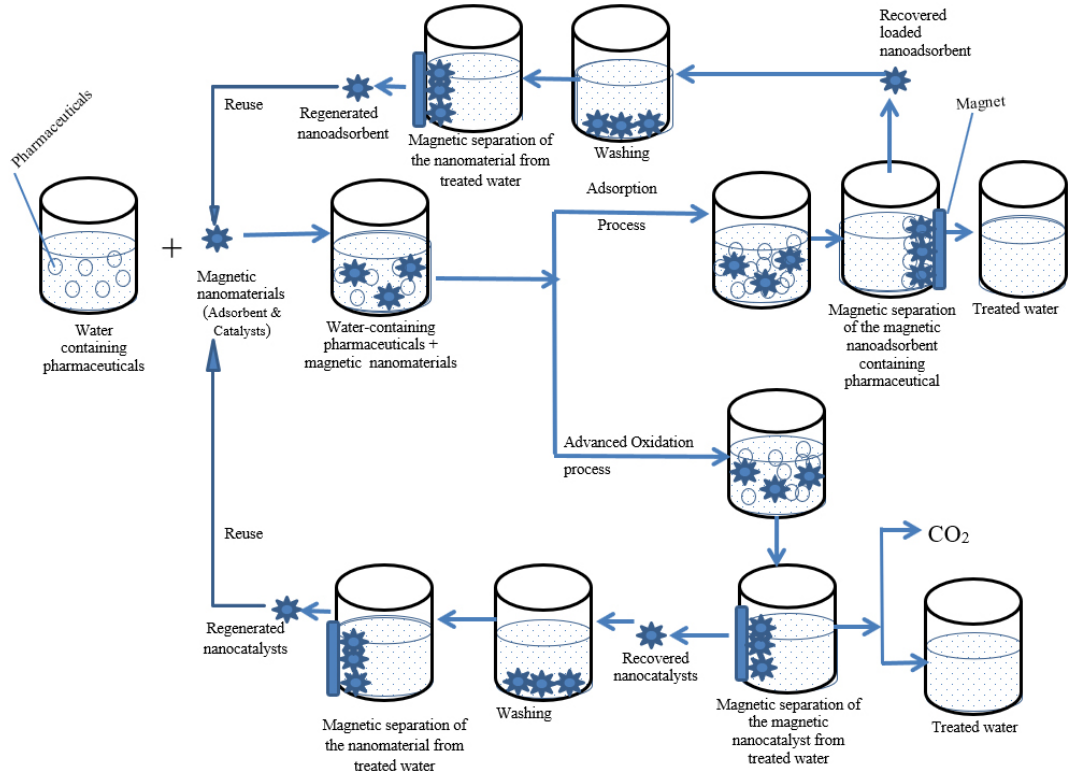

Figure 6. Schematic representation of removal of pharmaceuticals from water resources through adsorption and advanced oxidation processes by magnetic iron-based nanomaterials

7.1. Removal of pharmaceutical by adsorption process

Adsorption is a widely used technique for removing pharmaceuticals from water resources. Magnetic iron-based nanomaterials have a high surface area and strong magnetic properties, making them excellent adsorbents for removing pharmaceuticals from water resources, Table 1. The adsorption process involves the attraction of pharmaceutical molecules to the surface of magnetic iron-based nanomaterials. The pharmaceuticals then bind to the surface of the magnetic iron-based nanomaterials, which can be easily removed from the water using a magnetic field. The mechanism of removal of pharmaceuticals by adsorption process using magnetic iron-based nanomaterials is based on the principle of the Van der Waals forces, pore filling/size-selective adsorption, electrostatic interactions, hydrogen bonding, π-π interaction/stacking hydrophobic interaction (Zhuang, Chen, *et al.*, 2020; Zhuang, Cheng, *et al.*, 2019; Zhuang, Liu, *et al.*, 2019, 2020; Zhuang, Zhu, *et al.*, 2020). Several factors, such as pH, concentration, temperature, and contact time, influence pharmaceuticals' adsorption on magnetic iron-based nanomaterials, as in Table 1.

Various studies have shown that magnetic iron-based nanomaterials such as Fe_3O_4, Fe_2O_3, cysteine-modified silane-coated magnetic nanomaterial, Magnetic genipin-crosslinked chitosan/graphene oxide-SO_3H, Fe_3O_4-beta-cyclodextrin, and Fe_3O_4/GO etc., can effectively remove various pharmaceuticals such as ibuprofen, paracetamol, and tetracycline etc. from water resources, Table 1. Modifying the magnetic iron-based nanomaterials' surface with different functional groups such as carboxyl, amine, and hydroxyl can improve adsorption, as observed from several studies in Table 1.

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S N	Magnetiic Nanomaterials	Synthesis methods	Pharmaceuticals	Optimum Conditions	% Removal	Adsorption capacities (mg/g)	Detection technique	Reusability Cycle	Ref.
1	AC/Fe_3O_4	Co-precipitation	Ceftriaxone	pH; 3.14, contact time;90 minutes, Initial concentration; 10 mg/L, Adsorbent dosage; 1.99 g, Temperature; 298 K	97.18	28.93	HPLC/ UV-Visible Detector	6	(Yegane *et al.*, 2018)
2	$CoFe_2O_4$/AC@Ch	Co-precipitation	Ciprofloxacin	pH; 5, contact time; 15 minutes, Initial concentration; 10 mg/L, Adsorbent dosage; 0.1 g, Temperature; 298K	93.5	188.68	UV/Vis spectrophotometer	3	(Malakootian *et al.*, 2018)
3	Fe_3O_4/Biochar	Co-precipitation	Ibuprofen	pH; 8, contact time; 5 minutes, Initial concentration; 700 mg/L, Adsorbent dosage; 2.5 g, Temperature; 298K	99.2	39.9	UV/Vis spectrophotometer	5	(Liyanage *et al.*, 2020)
4	Magnetic graphene-anchored zeolite imidazolate (Fe_3O_4/ZIF-8-G)	Co-precipitation	Tetracycline	pH; 6, contact time; 10 hours, Initial concentration; 200 mg/L, Adsorbent dosage; 0.2 g, Temperature; 308 K	96.5	382.58 Freundlich	HPLC-MS	5	(Wang *et al.*, 2020)
5	Fe_3O_4@C	Sonochemical	Sulfamethoxazole	pH; 6.7, contact time; 24 hours, Initial concentration; 100 mg/L, Adsorbent dosage; 1.0 g, Temperature; 298K	89	1200	HPLC-MS	9	(Bao *et al.*, 2014)
6	Fe_3O_4@SiO2/SiTMC	Sol gel	Sulfamethoxazole	pH; 5, contact time; 24 hours, Initial concentration; 40 mg/L, Adsorbent dosage; 0.05 g, Temperature; 298K	NR	598	UV/Vis spectrophotometer	4	(Soares *et al.*, 2019)
7	Maghemite(γ-Fe_2O_3)	Precipitation/ Oxidation	Diclofenac	pH; 7, contact time; 24 hours, Initial concentration; 500 mg/L, Adsorbent dosage; 0.1 g, Temperature; 298 K	NR	252	UV/Vis spectrophotometer	5	(Oliveira *et al.*, 2017)
8	Magnetic poly (styrene-2-acrylamido-2-methyl propanesulfonic acid) (St-AMPS)	Sol gel	Diclofenac, DCF Ceftriaxone, CFX	pH; 4, contact time; 60 minutes, Initial concentration; 50 mg/L, Adsorbent dosage; 0.1 g, Temperature; 298K	DCF; 91.3 CFX; 94.5	DCF; 150.602 CFX; 119.904	UV/Vis spectrophotometer	4	(Hayasi & Saadatjoo, 2017)

S N	Magnetiic Nanomaterials	Synthesis methods	Pharmaceuticals	Optimum Conditions	% Removal	Adsorption capacities (mg/g)	Detection technique	Reusability Cycle	Ref.
9	Fe_3O_4@Cuttle-bone magnetic nanocomposite	Co-precipitation	Tetracycline	pH; 5, contact time; 90 minutes, Initial concentration; 8 mg/L, Adsorbent dosage; 0.2 g, Temperature; 298 K	80.15	14.94	HPLC/ UV-Visible Detector	NR	(Malakoo-tian & Shi-ri, 2021)
10	$NiFe_2O_4$	Sol gel	Oxycarbazepine	pH; 2.5, contact time 5 hour, Initial concentration; 0.144 mg/L, Adsorbent dosage; 0.6 g, Temperature; 303K	88.66	0.1034	UV/Vis spectropho-tometer	6	(Parashar *et al.*, 2019)
11	MFe_2O_4 (M = FeII, MnII, CoII, ZnII)	hydro-thermal	Tetracycline Oxytetracycline Chlortetracycline	pH; 6.9, contact time; 5 minutes, Initial concentration; 100 ug/L, adsorbent; 1.0 g	98.7	NA	UPLC-MS/ MS	9	(Bao *et al.*, 2013)
12	Fe_3O_4@C	Hyrother-mal	Tetracycline	pH; 5, contact time; 140 minutes, Initial concen-tration; 30 mg/L, adsorbent; 0.5 g, Temperature; 298 K	73.3	7.61	UV/Vis spectropho-tometer	NR	(Soares *et al.*, 2019)
13	ZIF-8@SiO2@Fe_3O_4	Sol gel	ceftazidime	pH; 6.3, contact time; 30 minutes, initial concentration; 1 mg/L, Temperature; 294.6 K	90	96.84	HPLC-Visible Detector	5	(Duan and Sun 2019)
14	Fe_3O_4-g-CN@ PEI-□-CD	Co-precipitation	Tetracycline	pH; 9.2, contact time; 20 minutes, Initial concentration; 265 mg/L, adsorbent; 0.008 g, Temperature; 317.1 K	98	833.33	HPLC-Visible Detector	NR	(Foroughi *et al.*, 2019)
15	-NH2-functionalized Fe_3O_4@SiO2 nanopar-ticles (MNPNH2)	Co-precipitation	Diclofenac	pH; 5, contact time; 6 hours, Initial concentration; 50 mg/L, adsorbent; 0.1 g, Temperatu-re; 298 K	97	565	UV/Vis spectropho-tometer	5	(Huang *et al.*, 2020)
16	Fe_3O_4/C	Hydro-thermal	ciprofloxacin	pH; 7, contact time; 3 hours, I initial concentration; 10 mg/L, adsorbent; 0.1 g, Temperature; 303 K	98.2	90.1	UV/Vis spectropho-tometer	5	(Mao *et al.*, 2016)
17	Magnetic nano-particle-decorated graphene oxide (GO-MNPs-SiO2)	Impregna-tion	Naproxen	pH; 5, contact time; 60 minutes, Initial concentration; 50 mg/L, adsorbent; 0.1 g	83 - 94	31.25	UV/Vis spectropho-tometer	18	(Nodeh *et al.*, 2018)

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S N	Magnetiic Nanomaterials	Synthesis methods	Pharmaceuticals	Optimum Conditions	% Removal	Adsorption capacities (mg/g)	Detection technique	Reusability Cycle	Ref.
18	Fe_3O_4/AC	Co-precipitation	Amoxicillin, Carbamazepine, Diclofenac	Adsorbent; 0.34 g, Initial concentration; 5 mg/L, Temperature; 333 K	70 77 84	NR	HPLC/ UV-Visible Detector	NR	(Rocha *et al.*, 2021)
19	$CoFe_2O_4$@methycellulose (MC)	Microwave assisted method	Tetracycline	pH; 6, contact time; 75 minutes, Initial concentration; 16 mg/L, adsorbent; 0.18 g, Temperature; 323 K	79.45	12.9	HPLC/ UV-Visible Detector	5	(Nasiri *et al.*, 2021)
20	Kaolinite supported $CoFe_2O_4$ (KCF)	Co-precipitation	Doxycycline	Contact time; 12 hours, Initial concentration; 100 mg/L, adsorbent; 0.15 g, Temperature; 333 K	68	400	UV/Vis spectrophotometer	3	(Olusegun & Mohallem, 2020)
21	Magnetic mesoporous carbon/β-cyclodextrin-chitosan (MMPC/CycChit)	Co-precipitation	Danofloxacin Enrofloxacin Levofloxacin	pH; 7, contact time; 30 minutes, Initial concentration; 10 mg/L, adsorbent; 0.36 g, Temperature; 298 K	98.7 99.1 96.8	130 195 165	HPLC-PDA	8	(Mashile *et al.*, 2020)
22	Fe_3O_4	Green	Sulfamethoxazole Trimethoprim Tetracycline Erythromycin Ampicillin Piperacillin tazobactam	pH; 5.5, contact time; 120 minutes, Initial concentration; 0.05 mg/mL, adsorbent; 0.1 g, Temperature; 303 K	90	0.9905 - 25.641	HPLC-DAD-MS	NR	(Stan *et al.*, 2017)
23	Ni0.5Zn0. $5Fe_2O_4$ magnetic nanoparticles	Co-precipitation	Diclofenac	pH; 3.2, contact time; 60 minutes, Initial concentration; 20 mg/L, adsorbent; 1.2 g, Temperature; 298 K	57.15	52.91	UV/Vis spectrophotometer	5	(Mohammadi *et al.*, 2021)
24	Magnetic genipin-crosslinked chitosan/graphene oxide-SO_3H	Impregnation	Ibuprofen (IP) Tetracycline (TC)	pH; 4, contact time; 120 minutes, IB; 10 mg/L, TC; 200 mg/L, pH 4, adsorbent; 0.5 g, Temperature; 313 K	NR	IB; 113.27 to 138.16 mg/g TC; 473.25 to 556.28 mg/g Freundlich	UV/Vis spectrophotometer	5	(Liu *et al.*, 2019)

S N	Magnetiic Nanomaterials	Synthesis methods	Pharmaceuticals	Optimum Conditions	% Removal	Adsorption capacities (mg/g)	Detection technique	Reusability Cycle	Ref.
25	Moringa oleifera Lam. seed husks functionalized with iron nanoparticles MOM-Fe_3O_4	Co-precipitation	Metformin	pH; 7, contact time; 1440 minutes, Initial concentration; 10 mg/L, adsorbent; 1.0 g, Temperature; 298 K	93.9	65.01 mg/g	UV/Vis spectrophotometer	NR	(Cusioli *et al.*, 2020)
26	Magnetic silica-based nanoadsorbent	Sol-gel	Ibuprofen (IBU) Diclofenac (DCF)	pH; 5.5, contact time; 24 hours, Initial concentration; 10 mg/L, adsorbent; 0.2 g, Temperature; 298 K	IBU; 68.7 DCF; 83	35	HPLC-UVD	8	(Peralta *et al.*, 2021)
27	Magnetic activated carbon-(AC-Fe_3O_4)	Co-precipitation	Promazine	pH; 8.5, contact time; 6 minutes, Initial concentration; 40 mg/L, adsorbent; 0.01 g	99.97	101.01	UV/Vis spectrophotometer	5	(D'Cruz *et al.*, 2020)
28	Fe_3O_4- beta-cyclodextrin	Co-precipitation	Naproxen (NAP), Carbamazepine (CBZ)	pH; 7, contact time; 120 minutes, Initial concentration; 20 mg/L, adsorbent; 0.1 g, Temperature 298 K	NAP; 80.2 CBZ; 75	NAP; 0.45 CBZ; 0.51,	UV/Vis spectrophotometer	3	(Ghosh *et al.*, 2013)
29	Powder activated carbon (PAC) combined with Fe_3O_4 magnetite nanoparticles (MNPs)	Co-precipitation	Amoxicillin	Temperature 20 °C, pH 5, 1 g adsorbent, concentration 50 mg/L and 90 minutes, pH; 5, contact time; 90 minutes, Initial concentration; 50 mg/L, adsorbent; 1 g, Temperature; 293 K	95.3	142.85	UV/Vis spectrophotometer	NR	(Kakavandi *et al.*, 2014)
30	Cysteine-modified silane-coated magnetic nanomaterial	Sol gel	Ibuprofen	pH; 6, Initial concentration; 50 mg/L, adsorbent; 30 mg,	82.9	138.1	UV/Vis spectrophotometer	3	(Kollarahithlu & Balakrishnan, 2019)
31	Fe_3O_4/GO	Co-precipitation	Chlorpheniramine	pH; 10, contact time; 24 hours, Initial concentration; 200 mg/L, adsorbent; 1.0 g,	NR	470	GC-MS	NR	(Li *et al.*, 2016)
32 mech	Magnetic graphene oxide	Co-precipitation	Tetracycline	pH; 4, Initial concentration; 50 mg/L, adsorbent; 0.19 g, Temperature; 298 K	78	174	UV/Vis spectrophotometer	5	(Li *et al.*, 2017)
33	$MgFe_2O_4$/gama-Fe_2O_3	Hydrothermal	Minocycline	pH; 7, contact time; 24 hours, Initial concentration; 100 mg/L, adsorbent; 0.025 g	NR	118.8	UV/Vis spectrophotometer	4	(Lu *et al.*, 2016)

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S N	Magnetiic Nanomaterials	Synthesis methods	Pharmaceuticals	Optimum Conditions	% Removal	Adsorption capacities (mg/g)	Detection technique	Reusability Cycle	Ref.
34	Fe_3O_4-MoO_3 and Fe_3O_4-MoO_3-AC	Impregnation	Ciprofloxacin	pH; 7, contact time; 60 minutes, Initial concentration; 20 mg/L, adsorbent; 0.05 g	Tap water; 95 Wastewater; 91 Seawater; 91.5	47. 294	UV/Vis spectrophotometer	5	(Mahmoud *et al.*, 2021)
35	Magnetic core-modified silver nanoparticles	Impregnation	Ibuprofen	pH; 7, contact time; 45 minutes, adsorbent; 0.07 g, Temperature; 298 K	93	NR	HPLC-UVD	4	(Vicente-Martínez *et al.*, 2020)
36	Magnetic nanoparticles coated zeolite Gama-Fe_2O_3-Zeolite	Co-precipitation	Diclofenac-Na (DCF), Naproxen (Nap), Gemfibrozil (Gem) and Ibuprofen (IB)	pH; 2, contact time; 10 minutes, adsorbent; 1.0 g	IB; 98.75 Nap; 99.7 DCF; 99.58 and Gem; 99.13	NR	HPLC-MS	NR	(Attia *et al.*, 2013)
37	Lignin-based magnetic nanoparticle adsorbent (LMNA)	Green	Diclofenac	pH; 5, contact time; 720 minutes, Initial concentration; 10 mg/L, adsorbent; 0.05 g, Temperature; 320 K	80	106.4	UV/Vis spectrophotometer	5	(Ye *et al.*, 2021)
38	Eggshell membrane (ESM)-derived $MgFe_2O_4$	Thermal decomposition	Doxycycline	pH; 5, adsorbent; 0.05 g, Temperature; 298 K (TC)	96	308.51	UV/Vis spectrophotometer	4	(Li *et al.*, 2017)
39	Fe_3O_4@graphene (Fe_3O_4@G)	Co-precipitation	Oxytetracycline (OTC) and tetracycline	pH; 7, Initial concentration; 1 mg/L, adsorbent; 0.06 g	Greater than 89	OTC; 336 TC; 423	HPLC-UVD	10	(Zhang *et al.*, 2017)

NR; Not reported

Table 1. Adsorption of Pharmaceuticals by magnetic nanomaterials.

7.2. Removal of Pharmaceutical by Advanced Oxidation Process

Advanced Oxidation Processes (AOP) are technologies that generate reactive oxygen species such as hydroxyl radicals (OH•) or sulfate radicals for oxidation of traceable organic contaminants or some inorganic pollutants or to increase wastewater biodegradability as a pre-treatment condition before biological treatment (Deng & Zhao, 2015; Wang & Xu, 2012). There are two advanced Oxidation Processes which are dark and light-driven. The dark advanced oxidation process includes; Ozone (O3), Fenton (Fe^{2+} + H_2O_2), Electrolysis (electrodes + current), and Sonolysis (Ultrasounds). The light-driven advanced oxidation process includes; Photolysis (UV + H_2O_2), Photocatalysis (light + catalyst) and Photo-Fenton (solar light + Fenton). Mechanisms of AOPs (Kommineni *et al*., 2008) are 1) the Formation of oxidants (e.g. hydroxyl radicals), 2) the reaction of these oxidants with organic compounds in the water-producing biodegradable intermediates, and 3) the reaction of biodegradable intermediates with oxidants referred to as mineralization (i.e. production of water, carbon dioxide and inorganic salts).

In this process, magnetic iron-based nanomaterials are added to the water, and then a reactive species is generated, such as hydrogen peroxide or ozone (Liu *et al.,* 2021; Liu & Wang, 2023; Tang & Wang, 2018; Wang & Tang, 2021). These reactive species then react with the pharmaceuticals, breaking them down into simpler, non-toxic compounds. The magnetic iron-based nanomaterials play a critical role in this process, as they help to generate and distribute the reactive species throughout the water. Several AOPs such as photocatalysis, photo-fenton, Fenton-like system, peroxymonosulfate (PMS), Sono-Fenton system, Persulfate (PS) system, UV/chlorine system, and Peroxydisulfate (PDS) system using different magnetic iron-based nanomaterials were investigated for degradation of pharmaceuticals, as shown in Table 2. The use of magnetic iron-based nanomaterials in AOPs has shown high removal efficiency and can be improved by modifying the magnetic iron-based nanomaterials' surface with different functional groups, Table 2.

8. RECYCLING AND REUSE EXPERIMENTS

Recyclability and reusability of magnetic nanomaterials are essential factors for practical application in water and wastewater treatment. To achieve recyclability and reusability, desorption experiments must be carried out using an appropriate desorbing agent at the end of each adsorption cycle. It is worth noting that some desorbing agents include ethano/water (Chen *et al*., 2018; D'Cruz *et al*., 2020; Hayasi and Saadatjoo 2017; Kakavandi *et al.,* 2014; Kumar *et al.,* 2018; Malakootian *et al*., 2019; Nasiri *et al*., 2019; Noroozi *et al.,* 2020; Soares *et al*., 2019; Tamaddon *et al*., 2020), distilled water (Dehghan *et al.,* 2018; Hou *et al.,* 2015; Jonidi *et al*., 2017; Kakavandi *et al*., 2016; Kumar *et al.,* 2018; Niu *et al*., 2011; Xiang *et al*., 2020; Yegane *et al*., 2018), dilute HCl solution (Jang *et al*., 2020; Vicente-Martínez *et al*., 2020; Zhang *et al*., 2017), methanol/water (Bao *et al*., 2014; Liyanage *et al*., 2020; Parashar *et al*., 2019), dilute NaOH (Lu *et al*., 2016), dilute NaOH/methanol (Mahmoud *et al*., 2021; Nodeh *et al*., 2018), thermal degradation (Mashile *et al*., 2020; Solís *et al*., 2021), ultrasonication (Ghosh *et al*., 2013), and water/acetone (Li *et al*., 2016; Stan *et al*., 2017). Furthermore, another significant factor for practical application catalysts and adsorbents is separation. Thus, magnetic nanomaterials are easily separated from the solution with the aid of an external magnetic field. All the publications reported the separation of the magnetic nanomaterials from the treated water or wastewater using an external magnetic field.

The percentage distribution of reuse and non-reuse of magnetic nanomaterials for the removal of pharmaceuticals from water was evaluated. About 78% of articles reported the reuse of magnetic nanomaterials for the removal of pharmaceuticals from water through adsorption and advanced oxidation processes. Most of the reusability studies were carried out between three and four cycles without the materials losing their efficiency. However, despite the fact that the magnetic nanomaterials were easily separated, 22% of the literature did not carry out reusability. This could be probably due to the researchers' interest since the materials were recovered.

S N	Magnetic Nanomaterials (Catalysts)	Synthesis methods	Type of Advanced Oxidation Processes	Pharmaceuticals	Optimum Conditions	% Removal	Detection Technique	Reusability Cycle	Ref.
1	$CuFe_2O_4$@methyl cellulose (MC)	Microwave-assisted method	Photocatalysis $CuFe_2O_4$@MC/UV	Ciprofloxacin	pH; 7, Concentration; 3 mg/L, Time; 90 minutes	Synthetic; 80.26 Real sample; 8072.87	HPLC-UV detector	4	(Tamaddon *et al.*, 2020)
2	nano$CoFe_2O_4$@methyl cellulose (MC)	Microwave-assisted method	Photocatalysis nano$CoFe_2O_4$@MC/UV	Metronidazole	pH; 11, Concentration; 5 mg/L, Time; 120 minutes, Catalyst; 0.2 g	85.3	HPLC-UV detector	4	(Nasiri *et al.*, 2019)
3	$ZnFe_2O_4$@carboxymethylcellulose(CMC)	Hydrothermal	Photocatalysis $ZnFe_2O_4$@CMC/UV	Ciprofloxacin	pH; 7, Concentration; 5 mg/L, Time; 100 minutes, Catalyst; 0.1 g	75 Synthetic and real water	HPLC-UV detector	5	(Malakootian *et al.*, 2019)
4	Fe_3O_4-graphene oxide	Co-precipitation	Fenton system Fe_3O_4-GO/H_2O_2 Peroxymonosulfate (PMS) system Fe_3O_4-GO/PMS peroxydisulfate (PDS) system Fe_3O_4-GO/PDS	Sulfamethoxazole, Norfloxacin, Tetracycline and Flumequine	pH 11, Concentration; 5 mg/L, Time 120 minutes, Catalyst 0.2 g,	83.3	HPLC-DAD detector	4	(Solís *et al.*, 2021)
5	$CuFe_2O_4$/GO	Co-precipitation	Peroxymonosulfate (PMS) $CuFe_2O_4$/GO/PMS	Metronidazole	pH; 5, Concentration; 30 mg/L, Time; 120 minutes, Catalyst; 0.2 g, PMS; 2 mM	100	HPLC-UV detector	5	(Noroozi *et al.*, 2020)
6	$CoFe_2O_4$	Co-precipitation	Peroxymonosulfate (PMS) system $CoFe_2O_4$/PMS	Ampicillin	pH; 7, Time; 25 minutes, catalyst; 0.1 g, PMS; 0.2 mM	90	HPLC-MS	5	(Balakrishnan *et al.*, 2021)

S N	Magnetic Nanomaterials (Catalysts)	Synthesis methods	Type of Advanced Oxidation Processes	Pharmaceuticals	Optimum Conditions	% Removal	Detection Technique	Reusability Cycle	Ref.
7	Maghemite nanoparticles	Co-precipitation	Photocatalysis Maghemite nanoparticles -UV/vis light	Tetracycline	Concentration; 83 ug/L, Time; 60 minutes, 10 mg catalyst; 10 tetracycline.	40	UV-vis spectrophotometer	NR	(Olusegun *et al.*, 2021)
8	Ag-$CuFe_2O_4$@WO3	Chemical deposition	Photocatalysis Ag-$CuFe_2O_4$@WO3/UV	Gemfibrozil (GEM), Tamoxifen (TAM)	pH; 5, Concentration; 5 mg/L, Time; 150 minutes, Catalyst; 0.2 g	GEM (81%) TAM (83%)	UV-vis spectrophotometer	5	(Sayadi & Ahmadpour, 2021)
9	$CoFe_2O_4$-GO	Hydrothermal	Peroxymonosulfate (PMS) $CoFe_2O_4$-GO/PMS	Norfloxacin	pH; 7, Concentration; 15 μM, Catalyst; 0.3 g, Temperature; 298 K	64.1	HPLC-UVD	4	(Chen *et al.*, 2018)
10	$CuFe_2O_4$@MC	Microwave-assisted method	Photocatalysis $CuFe_2O_4$@MC-UV	Ciprofloxacin	pH; 7, Concentration; 3 mgL, Tme 90 minutes, Catalyst; 0.2 g	80.74% (synthetic sample) and 72.87% (real sample)	UV spectrophotometer, HPLC-UVD (real wastewater sample)	4	(Nasiri *et al.*, 2019)
11	Fe_3O_4 MNP	Co-precipitation	Photo-Fenton system Fe_3O_4/ H_2O_2/Uv-vis	Ciprofloxacin	pH; 2.8, Concentration; 2.0 mg /L, Time; 1.8 minutes, H_2O_2; 2.50 mM, 500 W m^{-2}	85	UV-vis spectrophotometer	NR	(Lima *et al.*, 2014)
12	BiFeO3	Sol gel	Photocatalysis BiFeO3/visible light	Ciprofloxacin	pH; 6, Concentration; 1 mg/L, Time; 46 minutes, Catalyst; 2.5 g, Temperature; 303 K	100	UV/VIS Spectrophotometer	NR	(Mostafaloo *et al.*, 2020)

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S N	Magnetic Nanomaterials (Catalysts)	Synthesis methods	Type of Advanced Oxidation Processes	Pharmaceuticals	Optimum Conditions	% Removal	Detection Technique	Reusability Cycle	Ref.
13	Fe_3O_4@C	Co-precipitation	Photo-Fenton system Fe_3O_4@C/H_2O_2/UV	Tetracycline	Concentration; 20 mg/L, Time; 50 minutes, Catalyst; 0.15 g, H_2O_2; 3 mM	99	HPLCUVD	5	(Kakavandi *et al.*, 2016)
14	Humic acid coated Fe_3O_4 magnetic nanoparticles (Fe_3O_4/HA)	Co-precipitation	Fenton system Fe_3O_4/HA/H_2O_2	Sulfathiazole	Fe_3O_4 (3 g) and 10 mg/L of free HA or Fe_3O_4 (3 g) presoaked with 10 mg/L of HA, Time; 120 minutes	95	HPLC-PDA	3	(Niu *et al.*, 2011)
15	Fe_3O_4/Mn_3O_4	Impregnation method	Fenton system Fe_3O_4/Mn_3O_4/H_2O_2	Sulfamethazine	pH; 3, Concentration;20 mg/L, Catalyst; 0.5 g, Temperautre; 308 K, H_2O_2; 6 mM	94	HPLC-DAD	5	(Wan & Wang, 2017)
16	$ZnFe_2O_4$	Hydrothermal	Photo-Fenton system $ZnFe_2O_4$/H_2O_2/vis light	Tetracycline	pH; 4, Concentration; 40 mg/L, Time; 40 minutes, Temperature; 313 K, H_2O_2; 40 nmM	94.2	HPLC-MS	5	(Xiang *et al.*, 2020)
17	Fe_3O_4 @α-MnO2	Hydrothermal	Persulfate (PS) system Fe_3O_4 @α-MnO2/PS	Ciprofloxacin	Concentration; 50 mg/L, Time; 90 minutes, Catalyst; 1.0 g, Temperature; 298 K, $Na_2S_2O_8$; 2.0 g	90	HP LC-UVD	5	(Zhao *et al.*, 2014)
18	Magnetic nanocomposite (ZnO@Fe_3O_4)	Precipitation	Sonocatalytic system ZnO@Fe_3O_4/US	Amoxicillin	pH; 3, Concentration; 10 mg/L, Time; 120 minutes, catalyst; 0.8 g, US power (60 W)	90	HPLC-UVD	5	(Dehghan *et al.*, 2018)

SN	Magnetic Nanomaterials (Catalysts)	Synthesis methods	Type of Advanced Oxidation Processes	Pharmaceuticals	Optimum Conditions	% Removal	Detection Technique	Reusability Cycle	Ref.
19	Fe_3O_4	Co-precipitation	Sono-Fenton system US/Fe_3O_4/H_2O_2	Tetracycline	pH; 3, Time; 60 minutes, US power (80 W), 150 mmol/L H_2O_2, Catalyst; 1.0 g	93.7	HPLC-UVD	3	(Hou *et al.*, 2015)
20	Fe_3O_4	Co-precipitation	Sono-Fenton system US/Fe_3O_4/H_2O_2	Levofloxacin	pH; 6, Concentration; 20 mg/L; Time; 150 minutes, Catalyst; 1.0 g, H_2O_2; 5.0 mmol/L; Ultrasound power, 195 W	99	HPLC-MS/MS	NR	(Wei *et al.*, 2015)
21	Magnetic titanium carbide (Ti3C2Tx) MXene	Co-precipitation	UV/chlorine system Magnetic (Ti3C2Tx)/UV/chlorine	Diclofenac	pH; 7, Concentration; 30 mg/L, Time; 90 minutes, Catalyst; 3 g, Chlorine concentration; 30 mg/L	100	HPLC-MS	4	(Jang *et al.*, 2020)
22	AC@Fe_3O_4	Co-precipitation	Persulfate (PS) system AC@Fe_3O_4/PS	Tetracycline	40 mM PS, 10 mg/L TC, 0.4 g/L catalyst and pH 3.0) for a period 180 min	99.8	HPLC-UVD	5	(Jonidi *et al.*, 2017)
23	BiOCl/g-C_3N_4/Cu_2O/Fe_3O_4	Co-precipitation	Photocatalysis BiOCl/g-C_3N_4/Cu_2O/Fe_3O_4/sunlight	Sulfamethoxazole	pH; 6.5, Concentration; 100 μM, Time (Xe lap); 60 minutes, Time (natural sunlight); 120 minutes, catalyst; 0.2 g, Temperature; 303 K	99.5 (visible light), 92.1 (natural sunlight)	HPLC-MS	5	(Kumar *et al.*, 2018)

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S N	Magnetic Nanomaterials (Catalysts)	Synthesis methods	Type of Advanced Oxidation Processes	Pharmaceuticals	Optimum Conditions	% Removal	Detection Technique	Reusability Cycle	Ref.
24	ZnO/ Fe_3O_4-Sepiolite	Co-precipitation	Photocatalysis ZnO/Fe_3O_4-Sepiolite/sola light	Ibuprofen	Concentration; 10 mg/L, Time; 8 0.25 g catalyst; 0.25 g, Temperature 311 K, 18 hrs	89	HPLC-UVD	3	(Akkari *et al.,* 2018)
25	$Zn_{1.0}Fe_{2.0}O_4$	Combustion	Fenton-like redox system $Zn_{1.0}Fe_{2.0}O_4$	Diclofenac	pH; 5, Concentration; 10 μM, Time; 60 minutes, catalyst; 0.17 g, Temperature; 333 K	90	HPLC-PDA	NR	(Al-Anazi *et al.,* 2020a)
26	g-C_3N_4/TiO_2/ Fe_3O_4@SiO_2	Sol-gel	Photocatalysis g-C_3N_4/TiO_2/Fe_3O_4@ SiO_2/visible light	Ibuprofen	pH; 7, Time; 15 minutes, 330 W m^{-2},	97	HPLC-UVD	3	(Kumar, Khan, *et al.,* 2018)
27	$MnFe_2O_4$/biochar composite	coprecipitation	Photo-Fenton system $MnFe_2O_4$/bio-char/ H_2O_2/visible light	Tetracycline	pH; 5.5, Concentration; 40 mg/L, Time; 2 hours, H_2O_2; 100 mmol/ L	95	UV-Vis spectrophotometer	4	(Lai *et al.,* 2019)
28	Multi-walled carbon nanotubes (MWCNTs) -$NiFe_2O_4$ (NiFe-CNT)	Hydrothermal	Photo-Fenton system (MWCNTs) -$NiFe_2O_4$ (NiFe-CNT)/H_2O_2/ visible light	Sulfamethoxazole	pH; 3, Concentration; 5 mg/L, Time; 2 hours, catalyst; 0.025 g, H_2O_2; 1 μL/mL	100	HPLC-UVD	5	(Nawaz *et al.,* 2019)
29	rGO-AgO/Fe_3O_4	Impregnation	Peroxydisulfate (PDS) system PDS/rGO-AgO/ Fe_3O_4	Acetaminophen, Ibuprofen,	pH; 4, Time; 160 minutes, Catalyst; 0.1 g, PDS; (10 μM, 1 mM)	99	HPLC-DAD	NR	(Park *et al.,* 2018)

S N	Magnetic Nanomaterials (Catalysts)	Synthesis methods	Type of Advanced Oxidation Processes	Pharmaceuticals	Optimum Conditions	% Removal	Detection Technique	Reusability Cycle	Ref.
30	Biochar-TiO_2 magnetic nano-composites	Solvothermal	Photocatalysis Biochar-TiO_2 magnetic/UV	Sulfadiazine (SDZ) Oxolinic acid	Time; 1 hour, catalyst; 0.1 g, SDZ (5 mg/L) and OXA (10 mg/L)	SDZ; 87 OXA; 98	HPLC-UV	NR	(Silva *et al.,* 2021)
31	ZIF-8/$MnFe_2O_4$	Hydrothermal	Photo- Fenton system ZIF-8/$MnFe_2O_4$/ H_2O_2/visible light	Tetracycline	pH 3, Time; 90 minutes, catalyst; 0.3 g, concentration; 10 mg/L, H_2O_2 50 mM	92	UV-vis spectrophotometer	5	(Wang *et al.,* 2020)
32	TiO_2/Fe_3O_4	Hydrothermal method	Photo-Fenton system $TiO_2/Fe_3O_4/H_2O_2$/UV	Tetracycline	pH; 7, Concentration 50 mg/L, Time; 60 minutes, Catalyst; 0.3 g, Temperature 298 K,	98	UV-vis spectrophotometer	5	(Yu *et al.,* 2019)
33	$CuFe_2O_4$	Sol gel	Fenton system $CuFe_2O_4/H_2O_2$	Amoxicillin	pH; 4, 50 mg/L, Time; 30 minutes, Catalyst; 90 mg, Temperature; 293 K,	99.27	HPLC-Visible Detector	5	(Amraei *et al.,* 2016)

NR; Not reported

Table 2. Advanced oxidation processes for degradation of pharmaceuticals by magnetic nanomaterials.

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9. REAL WATER SAMPLES APPLICATION

The composition of water resources samples such as lake, river, sea, groundwater, and wastewater, etc. differ largely from distilled water used in laboratory experiments. Hence, newly developed magnetic materials' capability must be demonstrated with environmental water samples to understand the feasibility of the materials before the pilot scale. From the articles reviewed, 78% of publications did not carry out real water sample analysis and these represent a large portion of the research. Thus, more research is needed for real water samples. The other portion that reported real water analysis is 22%. Though the portion is very small, results demonstrated that the magnetic nanoparticles were effective in removal of pharmaceuticals from river water (Attia *et al.*, 2013; Lai *et al.*, 2019; Stan *et al.*, 2017; Wang *et al.*, 2020), lake water (Huang *et al.*, 2020; Wang *et al.*, 2020; Zhang *et al.*, 2017), seawater (Attia *et al.*, 2013; Vicente-Martínez *et al.*, 2020), industrial wastewater (Attia *et al.*, 2013; Kumar *et al.*, 2018; Malakootian and Shiri 2021; Mashile *et al.*, 2020; Nasiri *et al.*, 2019; Olusegun and Mohallem 2020; Rocha *et al.*, 2021; Stan *et al.*, 2017; Vicente-Martínez *et al.*, 2020; Wang *et al.*, 2020; Ye *et al.*, 2021), medical wastewater (Wang *et al.*, 2020), urban wastewater (Solís *et al.*, 2021), pool water (Zhang *et al.*, 2017) and tap water (Lai *et al.*, 2019; Stan *et al.*, 2017; Vicente-Martínez *et al.*, 2020; Wang *et al.*, 2020; Zhang *et al.*, 2017). These indicate that magnetic nanomaterials have good prospects for water and wastewater treatment.

10. DRAWBACKS OF MAGNETIC IRON-BASED NANOMATERIALS

Magnetic iron-based nanomaterials have shown promise for the removal of pharmaceuticals from water (Abdel Maksoud *et al.*, 2020). However, it is important to consider several drawbacks associated with these materials. One limitation is that they may not be effective for the removal of all types of pharmaceuticals, especially those that are highly soluble and do not adsorb well onto the surface of the nanomaterials (Wu *et al.*, 2008). This can limit their overall effectiveness in water treatment applications. Another drawback is the cost associated with the production and functionalization of magnetic iron-based nanomaterials (Leonel *et al.*, 2021; Singh *et al.*, 2020). These processes can be expensive and may require specialized equipment and expertise, making the widespread implementation of these materials in water treatment systems economically challenging. Furthermore, there are concerns about the potential environmental impacts of using magnetic iron-based nanomaterials for water treatment (Guo *et al.*, 2013; Leonel *et al.*, 2021). While these materials are generally considered safe, their long-term effects on the environment are not well understood. Thorough studies are needed to assess any potential risks associated with their use. Additionally, the stability of magnetic iron-based nanomaterials in water is a concern (Aragaw *et al.*, 2021; Sharma *et al.*, 2015; Singh *et al.*, 2021). Over time, these materials can undergo degradation or aggregation, which can limit their effectiveness for water treatment. It is crucial to ensure that these materials maintain their stability and performance over extended periods to ensure their efficacy in water treatment applications.

11. SUMMARY, CONCLUSION AND FUTURE PROSPECTS

In this work, synthesis, characterization and application of magnetic nanomaterials in the removal of pharmaceuticals from water through adsorption and advanced oxidation processes were investigated. Several synthesis methods reported in the articles studied are Co-precipitation, sol-gel, hydrothermal, solvothermal, sonochemical, microwave-assisted, impregnation, thermal decomposition, combustion, chemical deposition, and green. However, co-precipitation was the most frequent synthesis method reported due to the following: 1) It is simple, 2) It can be carried out at an ambient temperature and 3) Scalable productions. The characterization techniques frequently reported in the evaluation of magnetic nanomaterials include SEM/EDX, XRD, FTIR, DRS/UV/VIS, TEM, DLS, TGA, BET and VSM. Only three analytical techniques were used in the detection of the pharmaceuticals from the literature evaluated. These analytical techniques are gas chromatography, UV/Visible spectrophotometry, and high-performance liquid chromatography (HPLC) coupled with either UV or mass spectrophotometry (MS) as HPLC/UV/MS. Noteworthy, UV/Visible spectrophotometry was mostly used in pharmaceuticals' residual analysis after adsorption because the interest is just quantification, but in advanced oxidation processes, since degradation

products are of interest, HPLC/UV/MS was frequently used. Furthermore, in studies where real water analysis was conducted, HPLC/UV/MS was used regardless of whether the removal process was based on adsorption or advanced oxidation processes. This indicates the sensitivity and effectiveness of HPLC/UV/MS over UV/Visible spectrophotometry.

Overall, the application of magnetic nanomaterials in the removal of pharmaceuticals from water through the adsorption process and advanced oxidation processes was found to be promising. The magnetic nanoparticles were separated from the solution by external magnetic field, regenerated using various desorbing agents and reused for several cycles while maintaining its efficiency. Quite a portion of about 78 % of publications reported the reuse of the magnetic nanoparticles. Some of the desorbing agents reported in the works of literature are ethanol/water, methanol/water, acetonitrile/water, 0.1 M NaOH, 0.1 M HCl and acetone/water. Although only 22% of research was demonstrated with real water samples, the results were almost the same as the ones carried out using the synthetic solution. Although magnetic nanoparticles are excellent adsorbents and catalysts for removing pharmaceuticals from water, according to the results of this review, the following factors must be taken into account to enable large-scale water and wastewater treatments;

- Researchers need to make sure that more magnetic materials are developed, non-toxic materials are used for coatings and functionalization, and ecotoxicity testing is done from the manufacturing stage of nanomaterials to water treatment.
- Since batch methods are frequently employed, there is a need for continuous (column) treatment.
- For effective water treatment, real water sample treatment and a particular magnetic separator technology are required.
- Researchers should employ HPLC/MS as an analytical technique to quantify the residual levels of medications because of their sensitivity.
- Costs related to research initiatives should be estimated. A partnership between academics and NGOs, the government, businesses, or funding organizations would encourage the rapid actualization of magnetic nanoparticles for treating pharmaceutical-containing water.

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Competing Interests

No conflict of interest. ♦

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