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REVIEW

Review on Plant Mediated Green Synthesis of Magnetite Nanoparticles for Pollution Abatement, Biomedical and Electronic Applications

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In nature, the iron oxide is found in various forms. It is chemically mixed to form iron oxides (compounds). Magnetite has helpful uses in different areas, such as medicinal carriers, MRI-contracting agents, tumour therapies, industrial, laboratory dyes adsorption and wastewater treatment of toxic metals such as mercury and arsenic. Magnetic iron oxide nanoparticles are generated using various methods such as wet, dry or microbiological processes. The drawbacks of conventional nanoparticles including attrition and pyrolysis include a defective surface formation, poor efficiency rates, high development costs and high energy consumption. The first approach is the green biosynthesis of the nanoparticles in which the metal atoms are clusters. The organic compounds can both minimize and cover nanoparticles in the process of synthesis in green materials. In recent years, nanocarriers, especially for poorly soluble medicines, have received growing attention for oral chemotherapy. Early disease bacteria, biopsy, cells, DNA, glucose and viruses are identified by biosensing. While several basic characteristics have different advantages and potential for biomedical use of magnets of iron oxides, more toxicological research is required on as-synthesized magnet iron oxide nanoparticles with clearly defined requirements for toxicity assessment.

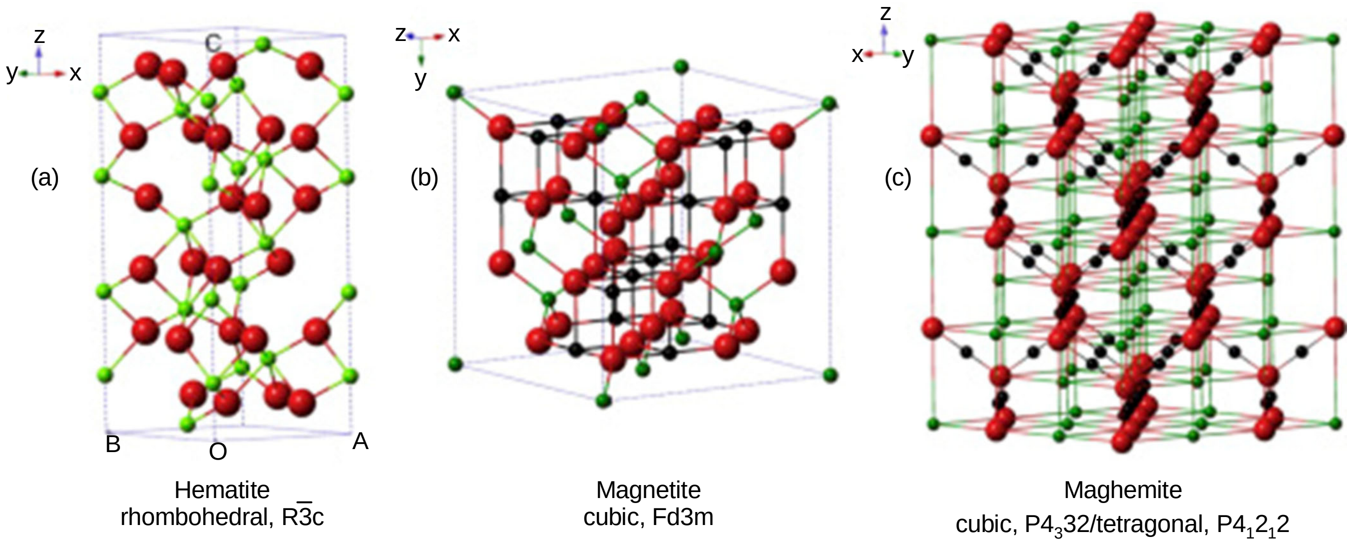
Keywords: Metal oxide nanoparticles, Magnetite, Hyperthermia, Biosensors.

INTRODUCTION

Nanotechnology can be characterized as the manipulation of matter through certain chemical and/or physical processes, which can be used in particular for materials that have special properties [1]. A microscopic particle with at least one dimension of less than 100 nm in size may be known as nanoparticles. They possess specific optical, thermal, electrical, chemical and physical properties, as opposed to bulk materials [2] and therefore have different applications in the areas of medicine, chemistry, the environment, electricity, agriculture, information, communications, the heavy industry and consumer goods [3].

Nano-metal oxides that are capable of taking on a large number of structural geometries with an electronic, metallic and semiconductor structure. The properties are optically, optoelectronically, magnetically, electrically, thermally, electrochemical, photo electrochemical, mechanically and catalytically [4]. In comparative to its bulk materials, the performance of the expected properties of metal oxides varies significantly with the reduction in particle size to nano and which is either increased, or totally new [5]. The most complex and significant kind of material is magnetic metal oxide that is attractive to research due to its unparalleled physical, chemical and structural properties.

In nature, the iron oxide exists in different forms. There are 16 types of iron oxides are available in nature among these three are popularly known are hematite (α-Fe_2O_3), magnetite (Fe_3O_4) and maghemite (γ-Fe_2O_3) in which magnetite has useful applications in various fields like medicine carriers, MRI contrasting agents, treatment of tumors [6] removal harmful industrial and laboratory dyes adsorption of harmful metals such as mercury, arsenic from wastewater treatment [7]. The crystal structures of hematite, magnetite and maghemite are shown in Fig. 1. Several physical properties along with magnetic nature of three iron oxide particles are listed in Table-1.



Hematite
rhombohedral, $R\bar{3}c$

Magnetite
cubic, Fd3m

Maghemite
cubic, $P4_332$/tetragonal, $P4_12_12$

Fig. 1. Crystal structure and crystallographic data of the (a) hematite, (b) magnetite and (c) maghemite (the black ball is Fe^{2+}, the green ball is Fe^{3+} and the red ball is O^{2-}) [8]

TABLE-1
VARIOUS PROPERTIES OF THREE OXIDES HEMATITE (Fe_2O_3), MAGNETITE (Fe_3O_4) AND MAGHEMITE (α-Fe_2O_3)

Property	Hematite	Magnetite	Maghemite
Phase	α-Fe_2O_3	Fe_3O_4	α-Fe_2O_3
Shape	Rhombohedral	Cubic	Cubic
Band gap	2.3 eV	0.1 eV	2.0 eV
Iron ions	Fe^{3+}	Fe^{2+}, Fe^{3+} (1:2)	Fe^{2+}, Fe^{3+}
Conductivity	n-Type semiconductor	n- and p-type semiconductor	n-Type semiconductor
Structure	Fe^{3+} occupy two thirds of octahedral	Cubic inverse spinel Fe^{2+} ions occupy half of the octahedral and Fe^{3+} evenly occupy remaining octahedral and tetrahedral	Fe^{3+} distributed in tetrahedral sites and octahedral sites
Magnetism	Weakly ferromagnetic	Ferri magnetism at room temperature change it to super paramagnetism, when particles below 15 nm	Ferri magnetism at room temperature

Iron oxide magnetic nanoparticles are prepared by various methods such as wet chemical, dry processes or microbiological techniques [9-14]. Briefly, iron oxide nanoparticles can be synthesized by three methods *viz.* (i) **Physical methods**: these are elaborate methods which are unable to regulate the nanometer size of particles [15]; (ii) **Chemical processes:** these are simple, tractable and efficient methods for managing the scale, composition and even the type of nanoparticles [16]. The co-precipitations of Fe^{2+} and Fe^{3+} iron oxides can be summed up with a base [17]. The structure, composition and size of the nanoparticles synthesized with iron oxide by chemical methods are determined by the salt type, ionic strength, pH and pH ratio of Fe^{2+} and Fe^{3+} [11]; and (iii) **Biological methods**: Conventional nanoparticles such as attrition and pyrolysis have disadvantages such as faulty surface formation, low performance rates, high production costs and a large energy demand [18,19]. Chemical synthesis methods (*e.g.* chemical reduction, sol gel, *etc.*) include the use of harmful substances, dangerous byproducts and precursor chemical pollution [19]. Clean, non-toxic and environmentally friendly nanoparticle synthesis procedures are therefore increasingly required. A very wide range of biological resources like microorganisms (bacteria, yeast, fungi, algae and viruses) and plants can be used for nanoparticle synthesis [20].

Plant mediated magnetic nanoparticles: The bottom up method is the green biosynthesis of nanoparticles in which the metal atoms form clusters and then finally nanoparticles. In green materials, the organic compounds can both reduce and cap the nanoparticles during the synthesis process. The size and form of the nanoparticles can be managed, which can be used for different applications. The simple method of synthesizing nanoparticles requires only materials like metal salt (precursor) and green substrates. The nanoparticles synthesis process allows various parameters, such as metal salt concentration, green substrate concentration, time and temperature to be modified and the solution's pH to achieve properties necessary for the respective applications. Green Fe_3O_4 nanoparticles biosynthesized may have better characteristics than physically synthesized Fe_3O_4 nanoparticles, such as higher biocompatibility and biodegradability. Due to the specific surface covering of green materials, they can therefore be used in biomedical applications which are non-toxic and biocompatible but can also be used in a targeted drug delivery with a position of Fe_3O_4 nanoparticles in some areas. It is possible to mitigate toxicity to the human body because green materials are healthy and thus advantageous to use to synthesize Fe_3O_4 nanoparticles for biomedicinal applications. In addition, Fe_3O_4 nanoparticles can be used with medications, enzymes or proteins that can be targeted by the assistance of external magnetic fields to tissues, organs, or tumors, or heated in alternating magnetic fields for the treatment of hyperthermia [21].

Synthesis of plant-mediated nanoparticles is the most effective way of generating large-scale nanoparticles within a short time. Fe_3O_4 nanoparticles can be synthesized across a wide range of effective studies, but the plant extract is the most widely used in green synthesis since large-scale processing, low-cost and environmentally friendly can be obtained easily [22]. Moreover, extracts from plants can be used in the synthesis of nanoparticles to reduce and stabilize nanoparticles due to phytochemical presence. The plant itself is composed of compounds of phytochemicals. They are expected to contain substantial quantities of phytochemical products such as flavonoids, xanthophylls, carotenoids, anthocyanins and phenolic acids. For synthesis there is also no need for additional surfactants or capping agents [23]. Metal ions are reduced to small nucleation centers in the aqueous salt solution. These nucleation centers are growing in size through the sequestration of more metal ions and the nucleation sites. The organic moieties in plant extract that endorse the capping of non-personal plant [24-26] are closely linked to these nanoparticles. Since the synthesized nanoparticles have a wide area to volume ratio, they have high surface energies. They are highly reactive and unpredictable. Capping nanoparticles prevents and stabilizes the agglomeration of nanoparticles [27,28]. At room temperature and pressure the entire process occurs quickly and easily in one stage. The synthesis prevents dangerous chemicals and toxic solvents from being required. Moreover, waste products can easily be disposed of in the atmosphere, as they consist mostly of the biomaterial of the plant. Quick, cost-effective, recreational and sustainable synthesis processes overall [25,29-31]. Furthermore, this approach can be used to achieve stable nanoparticles of desired size and morphology [32]. Therefore, biologically shaped nanoparticles have higher qualities than chemical nanoparticles. Tables 2 and 3 display the green materials used by different workers for Fe_3O_4 nanoparticles synthesis

Applications of magnetite nanomaterials

Environmental applications: Many of the world's most important challenges over the next few decades have to do with the quality of the atmosphere we live in. In all these areas, the use of magnetic nanoparticles is an increasingly important part of the many technologies and solutions currently being studied by scientists and engineers. The sector in which magnetic nanoparticles has to date made the greatest impact is possibly in the treatment of aqueous environments by eliminating industrial pollutants or enhancing the quality of drinking water source from groundwater and marine surroundings.

The changed behaviour is peculiar to the nanoscale, such as increased reactivities resulting from massive surface-to-volume ratio and magnetic phenomena such as super paramagnetic behaviour. Magnetic nanoparticles pair the modifications. With this effective combination, magnetic nanoparticles serve not only as efficient pollutant sorbents, but also for further processing and removal by magnet separate from the surrounding water media. Cutting and operation of magnetic nanoparticles produces a central shell structure that allows for remarkable selectivity and high sensitivity to particular chemical and metallic contaminants. Environmental chemosensors utilizing magnetic nanoparticles functionalized with chemical or fluorogenic molecules recently are receiving increased interest. These molecules also undergo a certain physico-chemical changes as a consequence of the selective capture of a target pollutant, which created a readable signal, which allows quick *in situ* detection. The iron reactivity can also be used to minimize contaminants to less toxic ingredients as nanoscale zerovalent iron (NZVI) to treat soil and groundwater pollution.

Chen *et al.* [73] functionalized boron nitride nanotubes with Fe_3O_4 nanoparticles showing effective As^{5+} removal over

TABLE-2
SYNTHESIS OF MAGNETITE NANOPARTICLES USING VARIOUS LEAF EXTRACTS

Plant extract	Shape	Size distribution (nm)	Ref.
Wedelia urticifolia (Blume) DC. leaf aqueous extract	Rod shape	15-20	[33]
Mentha pulegium L. leaves aqueous extract	Cubical shape	22-34	[34]
Aqueous leaf extract of *Zanthoxylum armatum* DC.	Spherical	17	[35]
Aqueous neem leaf extract	Nearly spherical	9-12	[36]
Aqueous green tea leaf extract	Spherical	25-30	[37]
Aqueous *Cynara cardunculus* leaf extract	Semi spherical	13-14	[38]
Aqueous pomegranate leaves	Nano rod	45-60	[39]
Aqueous lemon grass leaf extract	Clusters	~ 23	[40]
Aqueous Andean blackberry leaf extract	Spherical	30-78	[41]
Aqueous *Lagenaria siceraria* leaves extract	Cubic	30-100	[42]
Graptophyllum pictum leaf aqueous extract	FCC phase	~ 24	[43]
Moringa olifera leaf aqueous extract	Cubic	14-18	[44]
Myrtuscommunis L leaf aqueous extract	Cubic	10-12	[45]
Aqueous leaf extract of *Cynometra ramiflora*	Spherical	50-70	[46]
Eichhornia crassipes leaf extract	Rod shaped	15-20	[47]
Carica papaya aqueous leaf extract	Irregular	4-22	[48]
Azadirachta indica (neem) aqueous leaf extract	Ellipsoid	45-65	[49]
Aqueous extract of *Ficus hispida* L.	Spherical	10-12	[50]
Wrightia tinctoria aqueous leaf extract	Rhombic	105-145	[51]
Aloe vera plant aqueous extract	Spherical	~ 6-30	[52]

TABLE-3
SYNTHESIS OF MAGNETITE NANOPARTICLES USING VARIOUS PARTS OF PLANT EXTRACTS

Plant extract	Shape	Size distribution (nm)	Ref.
Aqueous potato extract	Cubic	38-42	[53]
Aqueous pod extract of *Dolichos lablab* L	Spherical	4-30	[54]
Brown aqueous seaweed (BS, *Sargassum muticum*)	Cubical	14-22	[55]
Aqueous fruit extract of edible *C. guianensis* (CGFE)	Spherical	17-80	[56]
Aqueous *Kappaphycus alvarezi* see weed	Spherical	11-20	[57]
Aqueous *Aloe vera* & flax seed extract	Spherical	30-50 & 30-40	[58]
Cnidium monnieri (L.) Cuss (CLC) seed aqueous extract	Spherical	35-45	[59]
Aqueous waste onion sheathing (*Allium cepa*)	Sheet	11-20	[60]
Plantain peel aqueous extract	Spherical	> 50	[61]
Acid-modified maize cob aqueous impregnated	Clusters	6-7	[62]
Aqueous *Syzygium cumini* seed extract	Spherical	9-20	[63]
Aqueous *Pisum sativum* peels extract	Spherical	20-30	[64]
Aqueous water melon (*Citrullus lanatus*) rind extract	Spherical	17	[65]
Aqueous soya bean sprouts extract	Spherical	~ 8	[66]
Satureja hortensis essential oil	Cubic	9-27	[67]
Persicaria bistorta root extract	Semi-spherical	45	[68]
Aqueous *Calliandra haematocephala*	Spherical	~ 8	[69]
Lathyrus sativus peel extract	Spherical	~ 18	[70]
Cynometra ramiflora aqueous fruit extract	Spherical	55-70	[71]
Aqueous plant extract of *L. camara*	Spherical	28	[72]

a wide range of pH values. Surface modification of γ-Fe_2O_3–Fe_3O_4 nanoparticles to enhance the removal of Cr^{6+} from wastewater was performed using a δ-FeOOH coating [74] and water-soluble polyethylenimine (PEI), which acted as a positively charged adsorbent that was effective at low pH [75]. Fe_3O_4 and $Fe_3O_4@SiO_2$ nanoparticles functionalized with 2-mercaptobenzothiazole [76], naphthalimide [77], mercapto polymers [78] and thiol [79] have all proved effective in the selective and sensitive adsorption and detection of Hg^{2+}. An interesting study by Farrukh *et al.* [80] outlined the use of 'polymer brushes' grown on Fe_3O_4 nanoparticles through a surface initiated polymerization process to demonstrate the complete removal of Hg^{2+} from water [80].

Shin & Jang [81] showed that Fe_3O_4 nanoparticles encapsulated in thiol-containing polymers are efficient at removing Ag^+, Hg^{2+} and Pb^{2+}. Gupta & Nayak [82] demonstrated an unusual and low-cost method of removing Cd^{2+} ions from simulated electroplating industry wastewater by co-precipitating orange peel powder obtained from agricultural waste with Fe_3O_4 with the resulting compounds showing a 82% removal efficiency. Zhang *et al.* [83] investigated the use of chitosan-coated octadecyl-functionalized Fe_3O_4 nanoparticles in the removal of a wide range of PFCs from environmental water samples from various locations determining detection limits between 0.075 and 0.24 ng L^{-1} with recoveries above 56%. Li *et al.* [84] used mixed hemimicelles composed of Fe_3O_4 nanoparticles coated with cetyltrimethylammonium bromide to remove chlorophenols from environmental water samples. A range of sulphonamide compounds have also been removed from water samples using mixed hemimicelles chemisorbed onto Fe_3O_4 nanoparticles [85]. A composite consisting of Fe_3O_4 nanoparticles supported on graphene sheets was used for the removal of both methylene blue and Congo red dyes [86]. The use of modified Fe_3O_4 nanoparticles to remove cationic dyes (crystal violet, methylene blue and alkali blue 6B) [87] and basic fuchsin, a magenta dye [88] has also been reported. Song & Gao [89] also showed that a photoactive TiO_2 shell surrounding a magnetic SiO_2-coated Fe_3O_4 core could be used as a magnetic photocatalyst, enabling the efficient degradation of methylene blue in aqueous solution with subsequent recovery of the nano-particle through magnetic separation. Magnetite nanomat-erials have been used for the removal of several toxic elements and dyes are literature listed in Table-4.

Biomedical applications: Nowadays, the successful treatment of various diseases and disorders greatly depends on drug delivery systems to create new and more efficient therapies. In this context, nanotechnology has emerged as a powerful strategy for the development of nanoparticles, with several biomedical applications for a range of diseases and infections from diagnosis to therapy. Several considerations must be taken into account before developing nano systems for biomedical applications. First of all, they must be made of biocompatible and biodegradable materials and provide sustained and controlled release of the bioactive agents as drug carriers [91]. Nanocarriers have attracted increasing attention in recent years for oral chemotherapy, particularly for poorly soluble drugs [92].

Attractive possibilities for use in biomedicine include magnetic nanoparticles. The living cells are 10 mm in size. Three orders of magnitude are smaller and their dimension is similar to that of viruses, proteins or genes, which means that they are "close" to the interest individual [93]. The particles may be guided or connected to particular areas of a body or cell when they are covered with unique biomolecules. In order to steer particles into specific regions in the body, these nanoparticles may also be manipulated by an external magnetic field. In addition, a resonating external magnetic field may be reacted by magnet nanoparticles. This allows energy to be transferred to the nanoparticles from the field. In recent years, therefore, a great deal of effort has been made to create magnetic nanoparticles and to understand their behaviour because of

TABLE-4
APPLICATIONS OF MAGNETITE NANOMATERIALS FOR THE REMOVAL OF SEVERAL TOXIC ELEMENTS AND DYES

Plant extract	Removal of dye/toxic metal	Dye removal (%)	Ref.
Cnidium monnieri (L.) Cuss (CLC) Seed extract	Cr(III), Pb(II)	88	[56]
Wedelia urticifolia (Blume) DC. leaf extract	Methylene blue	90	[33]
Potato extract	Methylene blue	88	[53]
Aqueous pod extract of *Dolichos lablab* L	Crystal violet	95	[54]
Waste onion sheathing (*Allium cepa*) Fe_3O_4@2D-CF	Arsenite	98.9	[60]
Acid-modified maize cob impregnated (16)	Methylene blue, COD reduction	99.63	[62]
Aqueous leaf extract of *Zanthoxylum armatum* DC	Methylene blue	70	[35]
Cynara cardunculus leaf extract	Methylene blue	97	[38]
Pomegranate leaves	Congo red dye	93	[39]
Pisum sativum peels extract	Methyl orange	96.2	[64]
Water melon (*Citrullus lanatus*) rind extract capped with DHPCT	Hg(II)	94	[65]
Leafs extract of '*F. chinensis* Roxb' and functionalized with 3-mercapto propionic acid	Malachite green, crystal violet and methylene blue	99.12	[90]

their unusual reaction to an external magnetic field. Magnetic nanoparticles are usually a magnetic core and a core that stabilizes the matter. Two forms can be structured to form the magnetic core: ferromagnetic crystallites in a nanoparticle aggregation and matrix shell dispersion in random ways. In order to ensure resistance and biocompatibility the shell is also used with an additional surface coating [94].

In biomedicine, it is also a benefit for the development of a concentrated specimen to isolate a special biological target structure from its natural environment. As a carrier are using the magnetic nanoparticles. Particles are protected by molecules with a structural affinity that is to be separated. The target structure should be binding with the nanopart during the incubation phase. An external magnetic field gradient is subsequently applied with a suitable magnetic separator and the entire magnet complex can be separated easily. The target structure is eventually isolated after the pollutants have been washed out [93].

Magnetic particle imaging: Superparamagnetic properties and large surface area useful for medical diagnosis can be seen on magnetic nanoparticles, such as Fe(III) oxide (Fe_2O_3) and magnetite (Fe_3O_4). Studies into the use of MRI magnetic nanoparticles showed great promise because of their superparamagnetic properties. A higher resolution non-invasive diagnostic test may lead to the use of these nanoparticles in MRIs. Those nanoparticles may also be used to label site of interest by attachment to biomarker or site(s) of interest of bioreceptors such as antibodies, aptamers, enzymes or proteins [95].

The new imaging technique first mentioned in 2005 is magnetic particle imaging [96]. The goal is positioned in an ongoing magnetic field with primarily saturated magnetization of the magnet nanoparticles. As a consequence, these particles do not have a harmonic signal. Only a small area is free of field and a harmonic signal can be observed from that particular area [97]. In medical applications such as the vascular or small intestinal imaging, macular pigment (MP) imagery has a high potential where quick dynamic information is needed and the targets lie relatively deep under the skin, this is because the signal is not affected by the interference tissue by magnetic particles.

Hyperthermia treatment: The theory that malignant cells are more heat sensitive than healthy tissue is built on hyperthermia. A local hyperthermia induced artificially can thus be used in the treatment of cancer [98]. The magnetic nanoparticles are scattered through the target tissue during hyperthermia treatment and an external magnetic field is applied with a certain field force and frequency. This causes particulate heating by neel loss, brown loss or loss of hysteresis [99]. Furthermore, the heat is dissipated in the surrounding tissue. Cancer tissues will be killed if the temperature reaches a therapeutic level of 42 ºC for 0.5 h or longer. The great benefit for such an application is that the targeted tissue is heated only while the other part is not affected [100], which is the magnetic nanoparticles used. In addition, the particles can be used to increase their absorption by binding with antibodies [101].

Biosensors: Biosensing is an effective medium for detecting early diseases of bacteria, biomolecules, cells, glucose DNA and viruses [102-105]. In the field of biomedicine, biological sensors are analytical instruments. Their primary purpose is the conversion into electric signals of biological, chemical or biochemical reactors [106,107]. Furthermore, magnetic nanoparticles (MNPs) surface functionalization is beneficial for the detection of molecular interactions; the vast surface area of MNPs allows the targeted interactions of bio molecules to work effectively [102,108]. By developing MNP-functionally Multi wall CNT-chitosan (MNP-FCNT-chitosan) and BSA compound films, Sun *et al.* [109] determined the use of composite immunosensors in carbofuran based on gold nanoparticles. In carbofuran detection, the immunosensors showed great stability, sensitivity and accuracy.

Electronic applications: The most popular, diverse and probably richest material groups in terms of physical, chemical and structural properties are the metal oxides among the different types of materials. For example, optical, optoelectronic, magnetic, electric, thermal, electrochemical, mechanical and catalyst properties are included in the abovementioned properties. In consequence of this, various applications for the use of metal oxides such as, for example, ceramics, (chemical, gas and bio-) sensors, actuators, lasers; waveguides; infra round and solar absorbers; pigments, photodetectors; optical switches; photochromics and refractory device; electro-catalyst and catalyst support (electro, photo, *etc.*).

In a process known as photocatalysis, the use of radiation (visible and ultraviolet), with a semiconductor material, has motivated many investigators to produce high-activity photocatalytic materials since the beginning of the 21st century [110,111]. A series of chemical reactions promoted by light is photocatalytic degradation. The production of catalysts that are easily regeneratable to fluid phase reactions including hydrogenation, aerobic oxidation, carbonylation, dehydrogenation and transesterification has recently been highly attracted by magnetic nanoparticles [112-117].

Conclusion

The size, dimensions and shape of magnetic nanoparticles of iron oxide are essential parameters for *in vivo* applications of pharmacokinetic and bio-distribution. Under different conditions, however, absolute control over the distribution of the shape and dimension of magnetic nanoparticles of iron oxide is still a problem and the different mechanisms for forming iron oxides have yet to be investigated [118]. Price, stability and compatibility are mostly significant advantages. Magnetic iron oxide nanoparticles are inexpensive to manufacture, have adequate physical and chemical stability and biocompatibility and are environmentally safe [119,120]. Although there are numerous specific properties of magnetic iron oxides, which have different advantages and possibilities for biomedical use, further toxicological research is needed on as-synthesized magnetic iron oxide nanoparticles, with clearly defined criteria for assessing toxicity [121,122]. The use of superior and faster methods for future studies could substantially encourage our understanding of the toxicity mechanisms of nanoparticle products [123]. In addition, magnetic oxide nanoparticles' biocompatibility is correlated with the intrinsic toxicity of functional layers and their biodegradation metabolites as well as with the reaction of the immune system after their administration.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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