A MODERN APPLICATIONS OF MAGNETITE NANOPARTICLES: ENVIRONMENTAL REMEDIATION AND BIOMEDICAL APPLICATIONS

Abstract

Nanotechnology has wide applications in medical science and for environmental friendly process. The co-precipitation method for the synthesis of magnetite nanoparticles are reviewed. Three factors which are temperature, pH and size plays an important role during this synthesis. Some of the future perspectives are presented in the next part of this review, which tells that iron-based nanoparticles can be used to generate green energy, Magnetite nanoparticles can be used for the detection and decontamination of inorganic and organic contaminants to counter the environmental pollution. The in vivo and in vitro applications in the biomedical world are enlighten. In particular, magnetite nanoparticles are widely used due to their chemical stability, high magnetic susceptibility, high saturation magnetisation. Magnetite shows superparamagnetic behaviour as its size is around 20nm, which is an important property for use in biomedical applications.

Synthesis

Synthesis of Magnetite

Co-precipitation method is one of the simplest and efficient method for the synthesis of aqueous phase [1]. A typical example of magnetite formation (Fe_3O_4) is described in equation [2]

$$Fe^{2+} + 2Fe^{3+} + 8OH^- \longrightarrow Fe(OH)_2 + 2Fe(OH)_3 \longrightarrow Fe_3O_4 \downarrow + 4H_2O$$

The co-precipitation method takes place mainly through a topotactic stage conversion root: akageneite stage (crystal nucleus formation) to goethite stage (narrow-shaped nanoparticle formation).

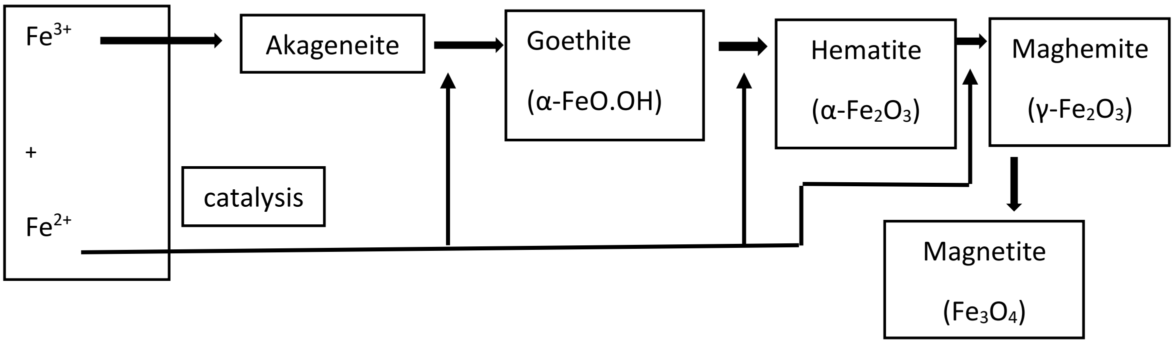

Figure 1: Formation root of MNPs via co-precipitation approach

Goethite (α-FeO·OH) is found in soils. The intermediate stage is the goethite stage [3]. From this stage magnetite converts into maghemite undergoes an oxidation process. Also, the annealing of maghemite converts it into hematite.

Iida et al. synthesised MNPs via hydrolysis in an aqueous solution consisting of Fe^{2+} and Fe^{3+} salts at varying ratios and 1,6-hexanediamine was utilised as the base [4]. When the ratios of Fe^{2+}: Fe^{3+} were increased, huge OH (hydroxide) particles were formed as the precursor of magnetite was promoted. When the diameter ranges from ~9 nm to ~37 nm the size of the MNPs increased. Also, by altering the ratio of Fe^{2+} and Fe^{3+} salts the magnetisation saturation value of the magnetite could be controlled.

During this synthesis, if the precipitation is below 60 °C it leads to the production of amorphous hydrated oxyhydroxide, which easily oxides into maghemite. The temperature should be above 80 °C to stimulate the formation of MNPs [5].

A suitable pH, excess amount of base addition was achieved for fast magnetite formation [6]. However, in this scenario, rapid formation of magnetite could also be attained with the usage of a strong base. [7]. By modifying the concentration of the base (e.g., NaOH) and the pH values the diameter of MNPs can be controlled.

At fixed pH:

Size of the MNPs α Concentration of NaOH.

At fix NaOH:

Size of the MNPs α 1 / pH values. So, at higher value of pH and lower concentration of NaOH, MNPs below the size of 3 nm were achieved [8].

FUTURE PERSPECTIVES OF MAGNETITE NANOPARTICLES (M-NPs)

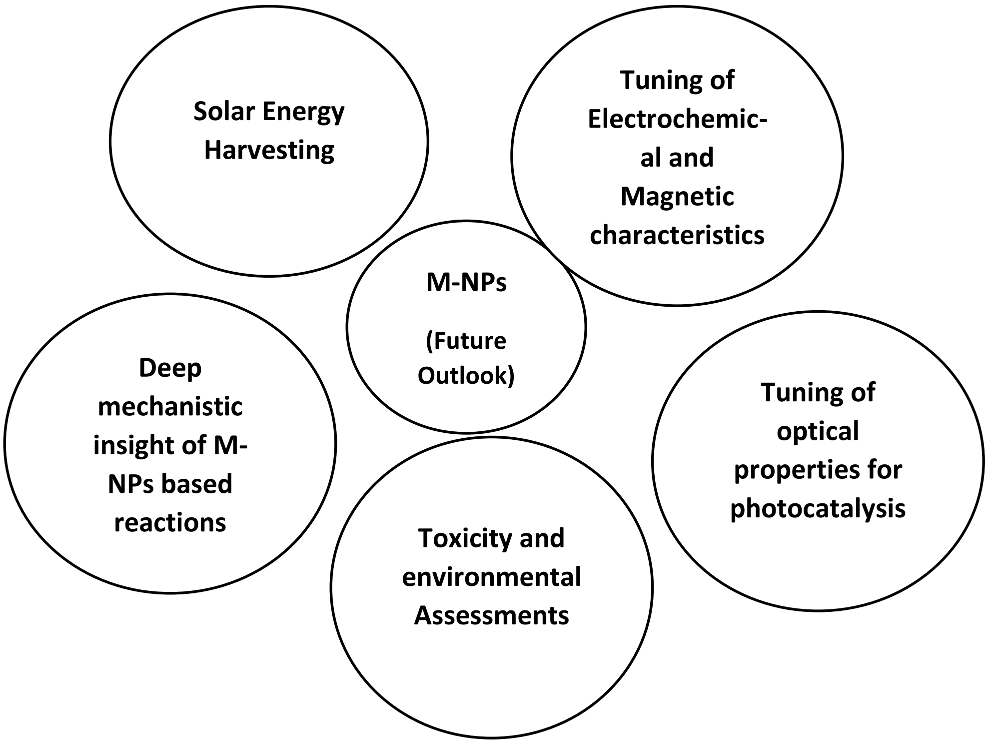

Figure 2: Future Perspectives for Magnetite Nanoparticles

ENVIRONMENTAL REMEDIATION BY MAGNETITE

Green and Renewable Energy Harvesting

To fulfil the energy requirements M-NPs are applied for renewable energy harnessing. It also substitute CO_2 emission with more reasonable and environmental friendly by-products. These NPs have been applied are discussed as follow.

CO_2 Remediation

Greenhouse gases, especially CO_2, continuously promote the greenhouse effect and hence create environmental misbalance. Therefore, to resolve this issue, researchers are trying to capture CO_2, convert it into CO, and lower and higher molecular weight hydrocarbons [9,10,11-12] via photocatalytic [13,14], electrocatalytic [11,15] and photoelectrocatalytic [16,17] conversion. M-NPs are found useful for CO_2 conversion due to adequate interfacial interaction of CO_2.

Spinel-shaped magnetite, having {001} and {111} facets, and its isostructural Fe_3S_4 are useful for the catalytic activation of CO_2, as they possess 2+/3+ mixed states. While the former is found to be significantly active in CO_2 activation, the latter showed an insignificant tendency toward the same [18]

Fossil Fuel Substituted Technologies for Sustainable Environment

For environmental sustainability Photovoltaic (PV) cells are best for the solar-induced energy harnessing to replace traditional fossil fuel consumption. Recently, PV technology gained significant attention due to dye-sensitized solar cells (DSSCs) [19]. A PV cell is composed of inorganic and organic counterparts. M-NPs-based electrodes are useful to complete the cell and to improve the overall energy generation capacity of the system [20-22]. Basically, DSSC consists of photoanode, electrolyte, the dye-sensitized counter electrode. Different modifications of anatase TiO_2 photoanodes were formerly utilized for DSSCs but due to high bandgap limits their efficiency [20,21,23]. Therefore, several semi- conductor counterparts were investigated for the substitution of TiO_2. M-NPs based materials which have smaller bandgap show decent photovoltaic performances in DSSCs.

Detection and Removal of Inorganic Contaminants

Use of heavy metals in industries has induce hazardous impacts. Researchers are trying to develop techniques which not only detect the ppm and ppb level of contaminants in the environment but also their removal. For the removal of copper (Cu(II)) [24], arsenic (As(III)) [25], mercury (Hg-(II)) [26], lead (Pb(II)) [27]etc. the effective use of iron oxide NPs has been widely reported. Magnetite acts as a good detector due to its dominant magnetic properties.

Magnetic ionic oxide coated NPs cross-linked to glyoxal and chitosan (Fe_3O_4/glyoxal/chitosan) absorbs 80−90% of toxic Cr(VI) from water [28]. At low pH (∼4), toxic Cr(VI) exists as $HCrO^{4-}$and reacts electrostatically with the NH^{3+} group on chitosan. The complex can be separated by the application of an external magnet, and the magnetite can be reused for further separation[28].

Similarly, 3-aminopropyl triethoxysilane (APTES) coated magnetite removed very toxic metals such as Cu(II), Cd(II), Ni(II), and Pb(II). The pH plays an important role in magnetite recyclability. when the pH reached 4, most magnetite was recovered [29]. The covalent thiosalicyl hydrazide/Fe_3O_4 magnetic nano adsorbents detect Cu(II), Zn(II),Pb(II), Cd(II), and Co (II) from industrial wastes with adsorption capacities of 76.9, 51.3, 188.7, 107.5, and 27.7 mg/g, respectively. These nanosorbents are reusable and environmental friendly [24].

Fan et al. reported the use of magnetic Fe_3O_4/chitosan NPs (Fe_3O_4/CS NPs) for the efficient removal of heavy metal ions, i.e., Pb(II) and Cd(II) (79.24 and 36.42 mg/g, respectively), from the aqueous solution.

Detection and Removal of Organic Contaminants

Organic contaminants (OCs) consists of group of hazardous chemicals found in various items like water disinfection by products, personal care products, pharmaceuticals, etc. Their continuous discharge lead to malfunctioning of various ecosystems, including human health. Here presents different ways by which the OC wastes can be removed.

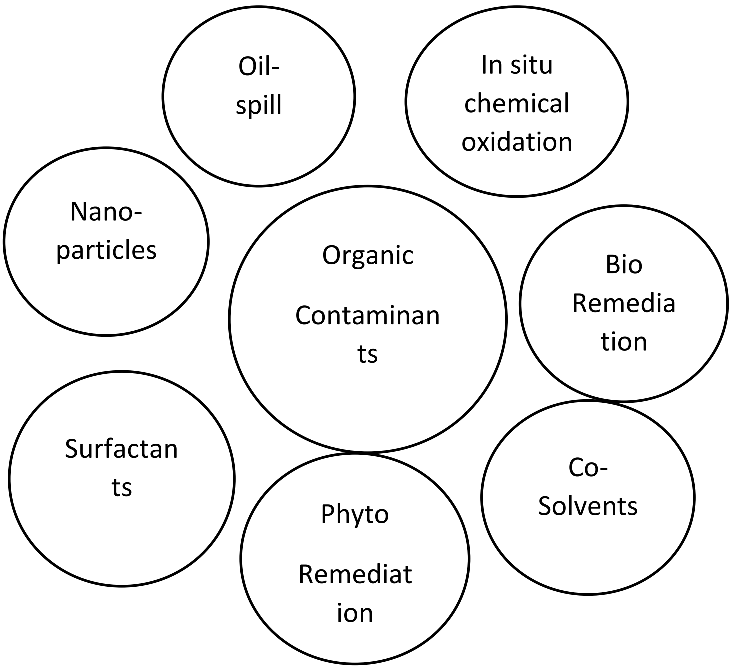

Figure 3: Common organic contaminants(OCs) remediation

Iron-based sensors are used for this purpose [30-34]. Azo dyes (reactive yellow and reactive black) are degraded by ferrous oxalate with surface-modified magnetite NPs in the presence of hydrogen peroxide [35].

Also Magnetite can effectively remove halo-genated organic compounds (HOCs) and polycyclic aromatic hydrocarbons (PAHs) [36]. Recently, adsorbents based on magnetic NPs in a silica mesoporous were found cost- effective and efficient for the removal of metal contaminants [1mg/L] and organic PAHs [1 mg/L]. The overall scheme is provided in Figure 4.

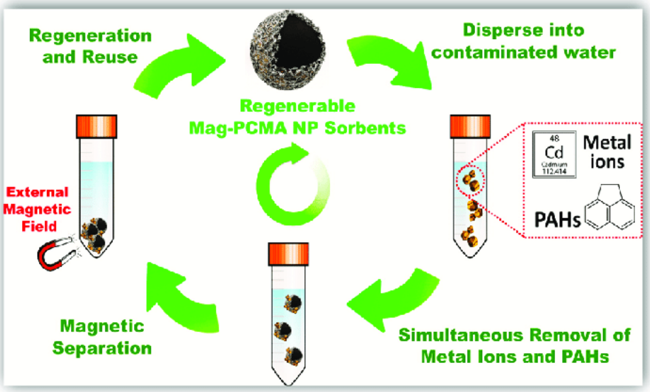

Figure 4: Removal of polycyclic aromatic hydrocarbon (PAH) OCs via magnetic NPs.

Another serious environmental problem is unwanted oil spills that disturb the ecological stability and destroy aquatic life. Modified asphaltene was used for the capping of magnetite, which disperse NPs, induces hydro-phobicity, and hence used for asphaltene separation. As a result heavy crude oil can be collected [37].

By a single-step solvothermal method, Shahid et al. successfully synthesized hydrophilic and hydrophobic iron oxide nanoparticles (SPIONs). The SPIONS functionalized with oleylamine (OLA) and polyethylene glycol (PEG-400) were found to highly stable in the Arabian seawater (ASW) [38] Figure 10 shows the demonstration of oil–water separation by using a magnet, while the graphs on the left show the T2 values.

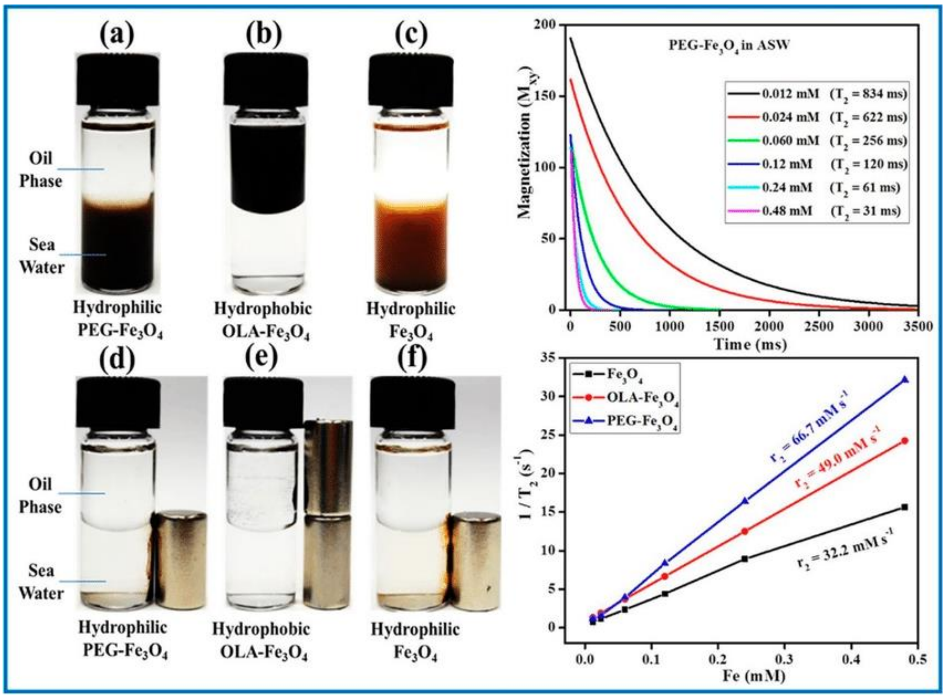

Figure 5: Functionalized magnetite NPs and their stability in oil-DI and oil sea water phases,with T_2 and $1/T_2$ relaxation curves.

Biomedical Applications of Magnetite (Fe_3O_4) Nanoparticles

MNPs have a great potential in the biomedical field (i.e., diagnosis). The biomedical applications of MNPs can be divided according to its applicability, they are inside the body (in vivo) or outside the body (in vitro). In vivo applications divided into diagnostic (MRI) and therapeutic (hyperthermia and TDD) applications. In vitro applications deal with magneto relaxometry and diagnostic separation, selection [6,39].

In Vivo Applications

Size and Surface modification are the two main factors in In vivo applications. In vivo can be classified into three categories; (i) magnetic vectors (ii) magnetic CAs in MRI; and (iii) magnetic hyperthermia [8]. Due to the advancement of technology in the biomedical field, there is a combination of in vivo applications simultaneously, like the usage of both TDD and MHT [40] and also MHT and MRI [41]. This section discussed about TDD, MRI, and magnetic hyperthermia.

TDD

Traditional TDD includes intravascular injection and chemotherapy, distribute anticancer drugs throughout the body, especially tumour tissues. Only a small amount of dosage reaches these tissues. But the toxicity of drug attack in healthy tissues are present. Therefore, TDD has been introduced as a replacement for the traditional chemotherapy treatment [42,43].

In cancer therapy, TDD has become the most advanced technologies. It focuses the loaded drugs on the cancer site, controls the amount of drug flow towards the tissues and reduces the side effects [42,44,45]. In TDD the application of MNPs has increased very much over the years [46]. MNPs are functionalised with polymers or gold. Functionalisation is done to check whether the anticancer drug could be loaded within the MNPs or conjugated on the surface of MNPs, as shown in Figure 6 [47]. Once the drug is loaded (drug-loaded MNP), an external magnetic field is applied to guide the drug-loaded MNP to the desired cancer site.

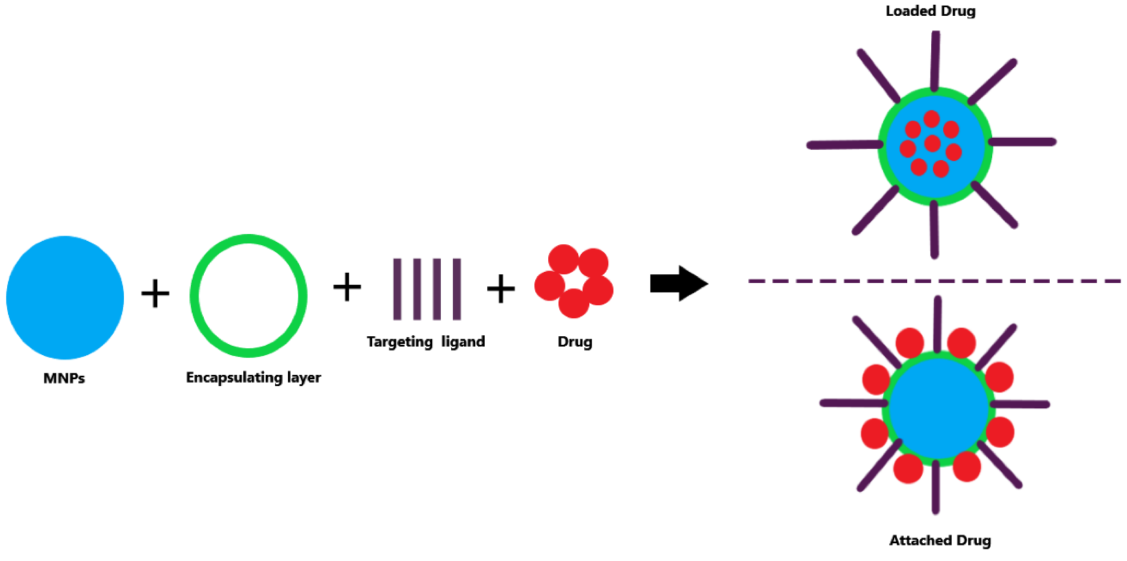

Figure 6: Illustration of drug loading roots in TDD.

MRI

In diagnostic medicine MRI is well-known biomedical imaging techniques. To generate high-resolution images of human tissues in 2D and 3D spaces it is applied. For the detection of pathological processes, it requires a CA to increase its sensitivity sharpness. MNPs are good and reliable CA in in vivo applications due to T1 and T2 relaxation time and a high proton magnetic moment alignment time that results in a much-improved MRI image [48]. When external magnetic field applied, MNPs produce gradients with large microscopic area. The microscopic field gradient reduces the relaxation time (T2*, T1 and T2), which results in a negative or dark contrast in T2-weighted images.

In the field of MRI, Specific cell labelling or cell tracking is another niche possessed by MNPs. When the cells are loaded with highly sufficient amounts of MNPs MRI provides resolution as small as the size of the cell

Gonzalez-Rodriguez et al. For MRI, magnetite nanocrystals are synthesised into hollow silicon nanotubes (SiNTs) as CA [49]. MNPs of 5-nm and 8-nm in average size were loaded into SiNTs of 40 and 70 nm-wall thicknesses (Figure 11). To attain colloidal stability, the nanocomposites were functionalised through an aminopropyl linkage with PEG-diacid (600).

Diagram

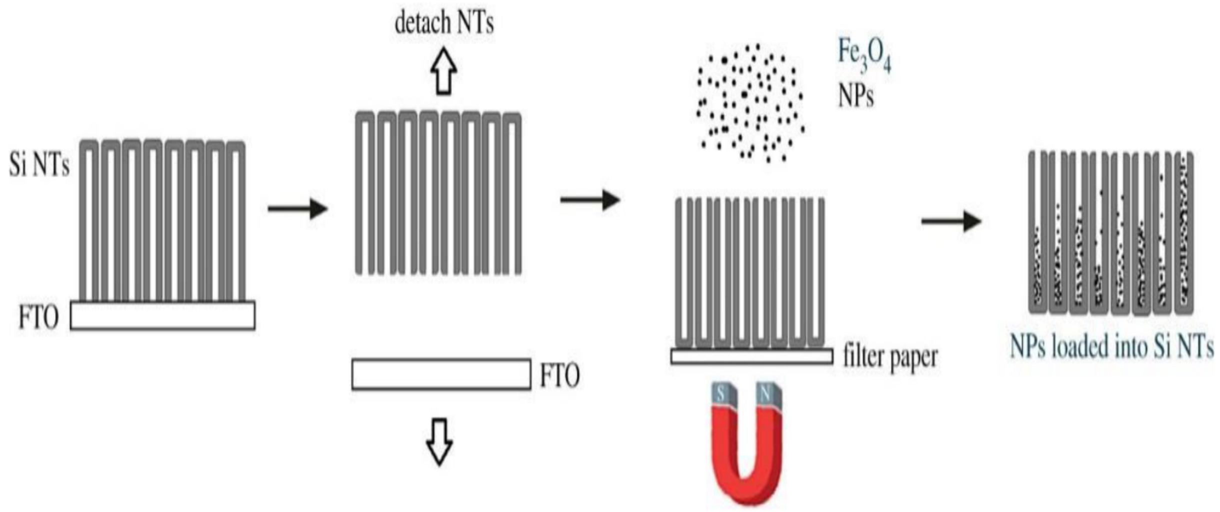

Figure 7: Schematic of MNPs being loaded into SiNTs

MHT

In cancer therapy Hyperthermia is another biological application of MNPs. It is a remedial approach where heat is induced to kill tumours [50,51]. MNPs act as thermal seeds. Once an external magnetic field is applied on MNPs, they heated up under a high frequency alternating magnetic field (AMF) at ~1 MHz [3] due to hysteresis loss. Due to higher heat of the cancer cells than the normal cells by not affecting the healthy ones, the temperature of the tumour cells is increased to 42–45 ◦C to destroy tumour tissues. When the heating reaches around 42–45 ◦C, the cancerous cells undergo cell apoptosis (i.e., hyperthermic effect). If the heating reaches >48 ◦C, the tissues undergoes necrosis (i.e., thermo-ablation) [42,50,51].

Local, regional, and whole-body hyperthermia are the three types of hyperthermia therapy. Local hyperthermia therapy focuses on a specific targeted site of the body part, regional hyperthermia therapy focuses on large tissue areas (e.g., limbs and organs) and the whole-body hyperthermia therapy treats with metastatic cancer which spread throughout the body. Figure 8 shows the process of MHT.

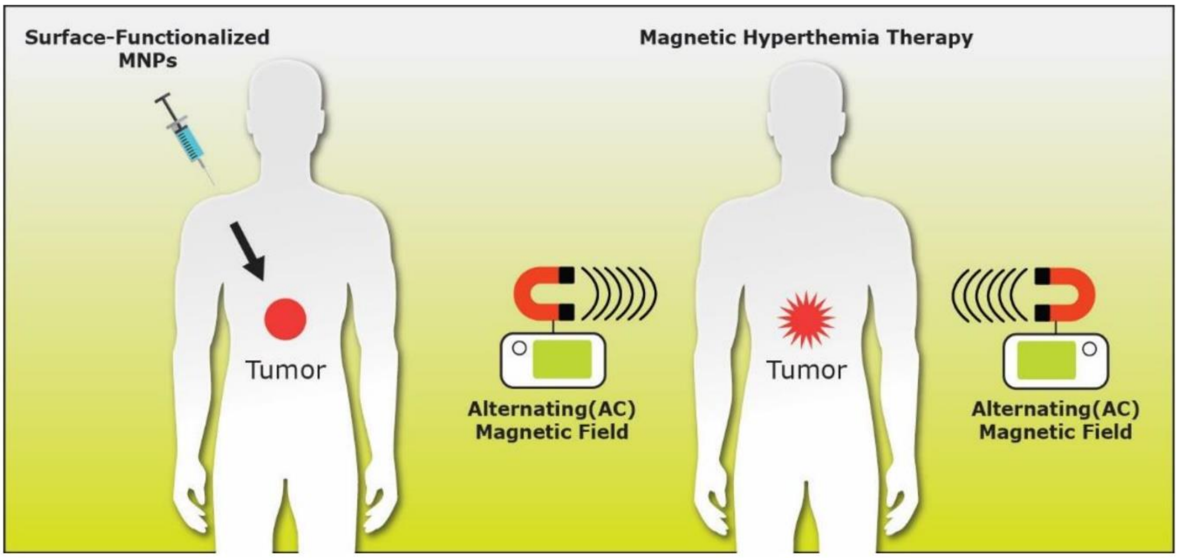

Figure 8: Illustration of typical magnetic hyperthermia therapy process.

In MHT, Superparamagnetic MNPs are the best materials. They are able to give a preliminary rise in temperature of 5 ◦C in just around 10 min [52].

In Vitro Applications

In vitro application is another type of MNP application. In this section, Bioseparation and Biosensors were discussed.

Bioseparation

MNPs can be successfully applied in Bioseparation because it is a crucial type of in vitro antibody, cell, DNA, enzyme, gene, bacterial and viral separation [53,54]. surface-modified MNPs with proper intermediates are normally used for surface enhacement. For surface functionalisation, ligands, polymers and surfactants are used to introduce functional groups (e.g., −COOH, −NH2, −OH and −SH) on the target biomolecules via selective adsorption [7].

On the surface of MNP@SiO2 at different pH levels and alkyl groups it is investigated the efficiency of separating bovine serum albumin (BSA) via functionalisation of hydrophobic pockets. By controlling the alkyl chain length, pH levels, salt concentration and size of the hydrophobic pocket they observed efficient magnetic separation and achieved strong magnetic separation [55].

Biosensors

For the detection of bacteria, biomolecules, cells, DNA, glucose, and viruses biosensing is an efficient platform [44,56-58]. In the biomedical field, Biosensors are analytical devices. Their main function is to convert biological, chemical, or biochemical response into electrical signals [59,60]. For the detection of molecular interactions, the surface functionalisation of MNPs is advantageous and for targeted biomolecule interactions the large surface area of MNPs permits efficient functionalisation [7,44].

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