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High values of the specific absorption rate of $NiFe_2O_4$-Chitosan nanostructures for magnetic hyperthermia

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***Abstract*—** Nickel ferrite nanoparticles is a kind of soft magnetic material that is one of the most attractive class of materials due to its appealing properties as well as various technical applications, such as being used as catalyst or in biomedical processes. The present paper focuses first on the synthesis of $NiFe_2O_4$ nanoparticles through co-precipitation method is reported and resulting in calcined nanoparticles that were achieved at different times and at constant temperature (773 k). Afterwards, they were dispersed in water that was incorporated by chitosan. Chitosan was bonded on the surface of nanoparticles. In order to assess the structural properties of nanoparticles, scanning electron microscopy (FESEM) with EDS and Vibrating Sample Magnetometer (VSM) were conducted at room temperature. FESEM were used to determine the size and shape of particle; the obtained results indicated that the particle was in the range of 15-40 nm and of a circular shape. A stable maximum temperature ranging from 30 to 42 was successfully achieved within 10 min. A specific absorption rate of up to 8.4 W/g was achieved. The study results revealed that the specific absorption rate (SAR) parameter of the coated nickel ferrite nanoparticle is more than that of pure nickel ferrite nanoparticles or Cobalt ferrite.

Keywords; Nickel ferrite nanoparticles, Chitosan biopolymer, Hyperthermia.

I. INTRODUCTION

Nickel ferrite nanoparticles ($NiFe_2O_4$) are known as one of the most important nanocrystalline spinel ferrites [1-5]. Used as permanent magnets and sensors, and employed in magnetic resonance imaging (MRI) enhancement, catalysis, magnetically guided drug delivery, and high density information strong technology are among the various applications of these materials.

Magnetic Nanoparticles are also employed for treating cancer. Surgery, chemotherapy, radiation therapy and hyperthermia are among cancer treatment options. The latter, clinical hyperthermia is of three broad categories, namely (1) localized hyperthermia, (2) regional hyperthermia and (3) whole-body hyperthermia. In this type of treatment, specific tissues or organs are heated (41–46 1C) for tumor/cancer therapy; the heat can be generated by radio frequency, microwave and laser wavelengths. The physical phenomenon of losses can be used to obtain the desired heat partially. In order to achieve the favorite temperature rise enhancement with low concentration of nanoparticles, it is desirable to have a high specific loss power (SLP) heat generated per unit mass of nanoparticles. In addition to field parameters, specific loss power of nanoparticles dispersions depend highly on particle size, size distribution, anisotropy constant, saturation magnetization, and surface modification.

$NiFe_2O_4$ is a cubic structure that is of an inverse spinel structure. Fe^{2+} and Ni^{2+} ions populate tetrahedral sites in this structure. Nevertheless, the strong dipole–dipole interaction causes the tendency of these $NiFe_2O_4$ nanoparticles to aggregation; hence, it is required to modify their surface with biocompatible and biodegradable polymer. As per the conducted literature review, starch, chitosan, polyvinyl alcohol, polyethylene glycol (PEG), oleic acid, dextran and lauric acid are the most appealing polymeric materials for surface coating of magnetic nanoparticles [5].

Chitosan is a polyaminosaccharide that is of many proper biological and chemical properties. It is not an expensive biopolymer and has reactive groups (OH and NH_2) which establish significant interactions with the surface of nanoparticles. In the present study, an attempt has been made to prepare the $NiFe_2O_4$ nanoparticles through co-precipitation method that has proved to be a proper technique for making small sized and mono-dispersity nanoparticles. And then, the suspension cross-linking technique was used for the preparation of chitosan coating on nanoparticles; glutaraldehyde was also used as

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a cross-linker. The nanoparticles were characterized by Vibrating Sample Magnetometer (VSM), scanning electron microscopy (FESEM) with EDS and due to the noticeable magnetic properties of $NiFe_2O_4$, the specific absorption rate (SAR) was calculated and compared to those in literature

II. MATERIALS AND METHODS

Firstly, 0.2 M $NiCl_2.6H_2O$ and 0.4 M $FeCl_3.6H_2O$ solutions were prepared separately. The solutions were then mixed with one another, and sodium hydroxide solution (3 M) was added dropwise to the final solution till pH was adjusted to around 13.

Oleic acid was added to the solution as a surfactant. And then, the temperature was increased up to 80°C for 40 min. Finally, the solution was centrifuged and washed with water and ethanol several times. The precipitation was dried in an oven at 80°C for several hours. Amorphous $NiFe_2O_4$ nanoparticles were obtained at this stage, and an additional process was used for obtaining the crystalline powder of nickel ferrite nanoparticles [1, 3].

Chitosan solutions of 0.5 wt% were prepared by dissolving the required amount of chitosan powder in 40 mL of a 2% acetic acid solution. 10 mL of the solution containing synthesized $NiFe_2O_4$ nanoparticles was poured into the prepared 0.5 wt% chitosan solution with vigorous stirring. 5 mL of a 25% NH_4OH solution was then added to the mixture. After the reaction, the chitosan coated nickel ferrite was washed with water, resuspended in 20 mL of a 0.5% acetic acid solution, and dispersed in a PBS solution by ultrasonication for 20 min.

III. RESULT AND DISCUSSION

Particle size, structure and magnetic properties of chitosan-coating of $NiFe_2O_4$ nanoparticles

Fig. 1 (a–b) shows the surface morphology of pure $NiFe_2O_4$ nanoparticles and chitosan coated $NiFe_2O_4$ nanoparticles. It can be observed in SEM and FESEM images that uncoated $NiFe_2O_4$ nanoparticles are highly agglomerated, while chitosan coated $NiFe_2O_4$ nanoparticles show that the $NiFe_2O_4$ nanoparticles are well-dispersed structures in the polymeric shell of chitosan. This may be due to the coating of chitosan on nanoparticles. Fig. 2 shows that EDS analysis of $NiFe_2O_4$ particles in figure 1 (b). It was also observed that the tendency of Magnetic saturation (Ms) to increase is consistent with the enhancement of crystallinity from 60 to 120 min; also the values of Magnetic saturation (Ms) decrease from 120 to 180 min due to the increase in particle size.

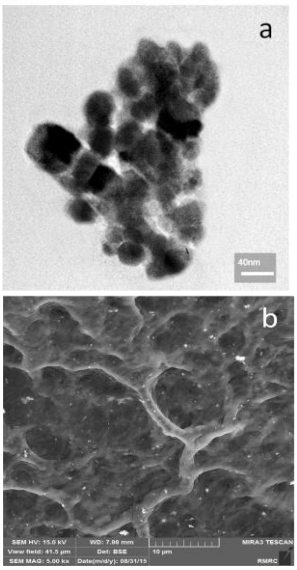

Fig. 1. SEM image of pure NiFe2O4 nanoparticles (a) and FESEM image of NiFe2O4 – chitosan (b)

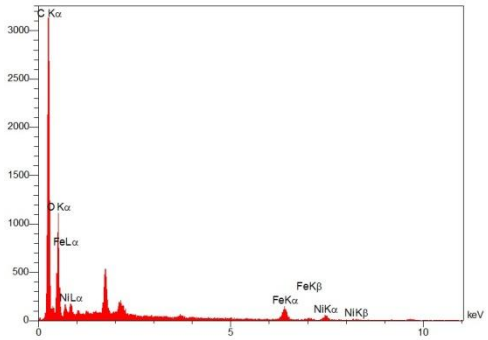

Fig. 2. EDS analysis of NiFe2O4 nanoparticles in figure 1 (b)

The increasing and decreasing filed sides (blanket) yielded a coercive force (Hc) of 90 (-95), 45 (-40) Orseted for the NiFe2O4 samples calcined for 120 and 180 min, respectively (Fig. 3) [2-4].

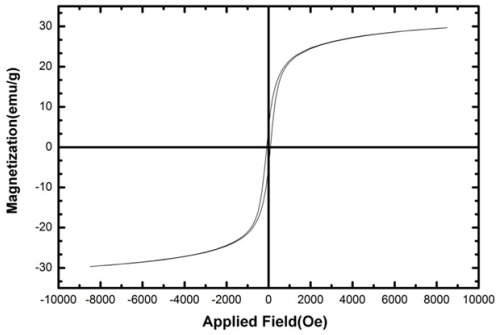

Fig. 3. The magnetic properties of NiFe2O4 – 2h of calcinations

Hyperthermia

Fig. 4 illustrates that $NiFe_2O_4$ nanoparticles (calcined for 120 min) are of higher magnetization as well as magnetic heating loss than other nanoparticles.

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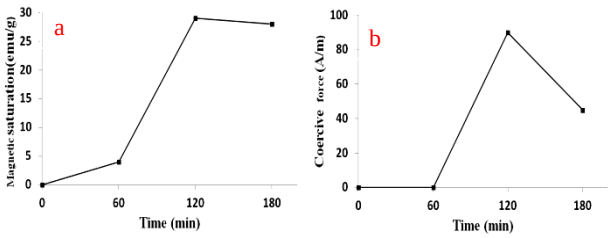

Fig. 4. Changes in specific saturation magnetization with the degree of calcination time (a) and changes in coercivity with the degree of calcination time (b).

Offering a moderate magnetic moment, chemical stability, and a high specific absorption rate (SAR) makes Nano-ferrites good candidates for hyperthermia purposes. The SAR was measured at room temperature using the heating yielded experimentally by the nanoparticles that calcined for 120 min (Fig. 5).

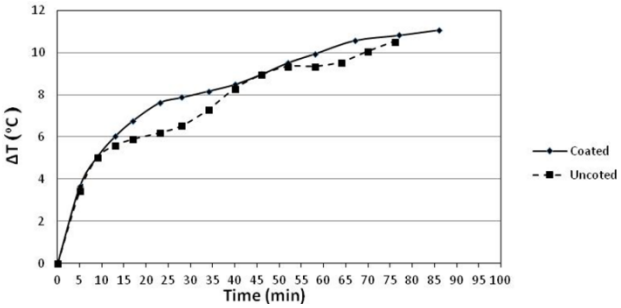

Fig. 5. Temperature increase of pure $NiFe_2O_4$ nanoparticles after 2 hours of calcination and $NiFe_2O_4$ - chitosan nanoparticles as a function of time

As shown in Table 1, $NiFe_2O_4$ chitosan coating nanoparticles, compared to those in previous studies, were of a reasonably high SAR at the physiologically expected frequency around 300 kHz.

Table 1. SAR of $NiFe_2O_4$ nanoparticles after 2 hours of calcination and $NiFe_2O_4$ - chitosan nanoparticles

Sample	**SAR (W/g)**	
	Uncoated	***With coating***
Calcined $NiFe_2O_4$	2.17	8.47

The attained SAR stood at about 3.5 and 6 for Cobalt ferrite at the same conditions. A remarkably small SAR was exhibited by uncoated $NiFe_2O_4$ nanoparticles, even at the same frequency. As shown in Figure 3, the temperature increased by an increase in time. The principal purpose of hyperthermia applications is generating a higher value of magnetic heat by a stable fluid in a lower exposure time. Ni-ferrite is of the highest level of SAR parameter in comparison with the other mentioned ferrites in biological applications (5 kA/m, 300 kHz).Thus, $NiFe_2O_4$ nanoparticles, for their high SAR at the physiologically tolerable range of applied magnetic fields and frequencies, can be a potential candidate as a clinical hyperthermia agent.

IV. CONCLUSIONS

Synthesis through co-precipitation was favourable for obtaining crystalline powders with nanosize nickel ferrite particles. SEM images confirmed that nanoparticles were dispersed uniformly (mono-dispersed), and were sphere in shape with a mean diameter of 18, 22, and 29 nm, respectively.

The results obtained from SEM indicate that $NiFe_2O_4$ nanoparticles were effectively coated by chitosan for various biomedical applications.

Calcined samples show that the size of particles increase by an increase in calcination temperature. High temperature calcinations also lead to an increase in the crystallinity of samples. It was verified that ferromagnetic $NiFe_2O_4$ nanoparticles were coated, and were of proper magnetic and structural properties; they were also of a high SAR for a hyperthermia agent application. Particularly, the adequately high heating temperature of the solid-state $NiFe_2O_4$ nanoparticles were readily controlled in the range of 30 – 45°C at the physiologically tolerable and biological safe range of the applied magnetic field; this allowed $NiFe_2O_4$ nanoparticles to be considered as a promising potential candidate as an in vivo hyperthermia agent.

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