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Core-shell Fe₃O₄@Ag magnetic nanoparticles detection using spin-valve GMR sensing element in the wheatstone bridge circuit

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Abstract

Core–shell Fe₃O₄@Ag magnetic nanoparticles (MNPs) integrated with a Wheatstone bridge–giant magnetoresistance (GMR) sensor provide access to GMR-based biosensors. The Fe₃O₄ nanoparticles synthesized using the coprecipitation method demonstrated 77 emu g⁻¹ of magnetization saturation (Mₛ), 51 Oe of coercivity (Hₑ), and particle size of 11 nm. Furthermore, core–shell Fe₃O₄@Ag MNPs prepared by the aqua-solution method possessed 53 emu g⁻¹ of Mₛ, 145 Oe of Hₑ, and 17 nm of particle size. This high Mₛ of nanoparticles not only offer a large induced magnetic field but is sufficient for particle penetration within the biofilms. It was discovered that the sensor can distinguish between the bare Fe₃O₄ with the Fe₃O₄@Ag nanoparticles through an output voltage increase corresponding to a decrease in Mₛ. The output signal of the sensor responds linearly to an increase in the core–shell Fe₃O₄@Ag nanoparticle concentration, owing to an increase in the induced-field. The sensor exhibits better sensitivity when applied in detecting less than 2 g L⁻¹ of nanoparticle concentration, that is, 0.76 mV per unit of concentration (g/L).

1. Introduction

In several fields, such as diagnostics, pharmacy, food safety analysis, and environmental monitoring, biosensing techniques, which are accurate, simple, affordable, and fast, and require a small sample, are indispensable. One promising technique for these applications is magnetic-based biosensing, in which magnetic particles are used as a label [1]. To obtain high sensitivity, the dimensions of the magnetic particle labeling must be comparable in size to biomolecules: a protein (5–50 nm) and a DNA (10–100 nm); thus, nanoparticles (NPs) are desirable. However, the detection of magnetic nanoparticles (MNPs) is a challenge because of their weak magnetic moment [2] owing to their limited physical volume, relatively large surface area, and thermal instability [3]. Therefore, a sensor capable of detecting a weak magnetic field (nT) at room temperature is required [4]. Giant-magnetoresistance (GMR) sensors are the magnetic sensors that meets these criteria [5, 6] with a spin-valve (SV) structure, which is based on the spin-dependent transport principle.

The basic rule of GMR sensors is detecting an induced magnetic field generated by the attached MNP labels on the sensing element surface [7]. Accordingly, the influence of the MNPs on the detection signals is a crucial aspect of the GMR biosensor [1, 8]. The GMR sensor offers other advantages. It has good compatibility for integration with other established electronic devices for real-time output [1]. Physically, this sensor has good thermal stability. Economically, the GMR sensor requires low power consumption [9] and low fabrication cost because of its compatibility with the existing large-scale machinery [1]. The absence of background noise in signal detection owing to the non-magnetic property of biomolecules [10] is also an advantage of this magnetic-based biosensor system.

Currently, Fe₃O₄ MNPs are investigated because of their potential utility in the biomedical field [11], including as a biomolecular label. The good saturation magnetization (Mₛ) of Fe₃O₄ MNPs, which offers not only a sufficient magnitude of the induced-field but also the flexibility of its magnetic moment control using an
external magnetic field, is an important feature in biosensor systems. The preferred method for large-scale synthesis of Fe₃O₄ MNPs is coprecipitation [12]. However, Fe₃O₄ MNPs synthesized by this method are susceptible to aggregation and oxidation [13, 14]. For biosensor applications, stable Fe₃O₄ MNPs are necessary [15]. To overcome this issue, the surface of the Fe₃O₄ MNPs needs to be coated with other functional materials to construct a core–shell structure with the Fe₃O₄ MNPs functioning as its core [13].

This surface modification method simultaneously expands the surface functionality of Fe₃O₄ MNPs [15] by binding to various types of biomolecules [16, 17] and preventing agglomeration and oxidation [16]. Various studies have been conducted to enrich the diversity of coating materials for various purposes. Materials that are commonly explored as functional surfaces of Fe₃O₄ MNPs are silica [18] and polymers [13]. Although they can prevent aggregation and corrosion of MNPs, they bind the bacteria [19]; thus, they are inappropriate for use in biomedical applications. Therefore, a new coating material that is more biocompatible is required. One material that is appropriate, but has not been widely explored as a functional surface of Fe₃O₄ MNPs, is silver (Ag) [15, 18].

Silver materials offer anti-bacterial activity [20, 21], good chemical stability, ease of functionalization with Fe₃O₄ [22], ease of functionalization via biomolecular amine/thiol terminal groups conjugation [15], and the ability to modify the magnetic properties of MNPs through surface interactions [23] such as Mₚ reduction [15, 16, 21]. The magnetic properties of the magnetic labels are crucial for the detection sensitivities [2] because of their effects on the induced-field and free-layer magnetization, resulting in a magnetoresistance change of the GMR sensor [10]. Nano-silver refers to plasmonic nanoparticles (PNPs) with typical localized surface plasmon resonance properties, which lead to strong optical absorption [24] owing to the mutual conduction electron response to light [15]. Hybrids of Fe₃O₄ as MNPs with Ag as PNPs in core–shell structures offer symbiotically improved functionalities [25] in a wide range of applications, such as magnetic and optical imaging, biosensors, cancer treatment, and bio-separation [24].

The superiority of the physical-chemical properties of the core–shell Fe₃O₄@Ag MNPs is attributed to their potential as a biomolecule label in GMR sensor systems. The Ag coating on the Fe₃O₄ core not only promises the wide application of the GMR sensor but also expands the MNP bio-functionality without sacrificing its Mₚ. Therefore, in this study, biocompatible core–shell Fe₃O₄@Ag MNPs were synthesized as a superior magnetic label. The effect of Ag on the magnetic core–shell on the response of the GMR sensor-based detection system and its response to the varied concentration of the core–shell Fe₃O₄@Ag MNPs were systematically investigated.

2. Materials and methods

2.1. Synthesis of core–shell Fe₃O₄@Ag Nanoparticles

The chemical coprecipitation method of ferrous and ferric salts in an aqueous ammonia solution was used to synthesize Fe₃O₄ MNPs [26]. All pure analytical materials were purchased from Merck (Darmstadt, GFR) and used without further purification. In brief, 8.109 g of FeCl₂·6H₂O and 4.170 g of FeSO₄·7H₂O, which were used as the Fe³⁺ and Fe²⁺ ion providers, respectively, were dissolved in 15 ml of de-ionized (DI) water separately under constant magnetic stirring speed (450 rpm) for approximately 15 min to attain a homogenous solution. Next, along with magnetic stirring (650 rpm) of both solutions for 15 min at approximately 60 °C, 24 ml of NH₄OH was dissolved in 36 ml DI water using a sonicator for 15 min. Subsequently, the NH₄OH solution was added to the ferrous and ferric mixture dropwise under constant stirring for 90 min at ±60 °C. During this drop, the color of the solution gradually transformed to solid black. The black precipitate was then separated from the basic solution using a permanent magnet and washed seven times consecutively using DI water to achieve neutral pH and eliminate impurities. After drying in the furnace for 2 h at 80 °C, the result obtained was a solid black powder of Fe₃O₄ MNPs, and it was then stored in a vacuum at ambient conditions for further use.

Core–shell Fe₃O₄@Ag MNPs were synthesized using the aqua-solution method [27]. To facilitate the positive charge of noble metal ion adsorption, the surface of the Fe₃O₄ MNPs that had been produced was modified using (3-Aminopropyl) trimethoxysilane (APTMPS) [24] purchased from Aldrich and used without further purification. In this modification, 500 mg of Fe₃O₄ MNPs was dissolved in 10 ml of APTMPS, 200 ml of ethanol, and 10 ml of NH₄OH by the sonification method for 90 min at room temperature to ensure that the entire surface of Fe₃O₄ was coated. After washing and drying, the surface-modified Fe₃O₄ MNPs were dissolved in 100 ml of ethanol and 80 ml of AgNO₃ (20 mm) under constant mechanical and magnetic stirring speeds (600 rpm) for 4 h at room temperature. The precipitate was then separated using a permanent magnet and washed thrice using ethanol. After drying in the furnace for 2 h at 60 °C, the solid powder of core–shell Fe₃O₄@Ag MNPs was finally obtained and then stored under vacuum in the ambient environment.

The magnetic performance and crystallinity of the MNPs were characterized using a vibrating sample magnetometer (VSM, Riken Denshi Co., Ltd.) and x-ray diffraction (XRD), respectively. Their size,
morphology, and elemental composition were characterized by a transmission electron microscope (TEM) in association with energy-dispersive X-ray spectroscopy (EDX).

2.2. GMR sensor design
In this work, an SV thin film with a structure of [Ta(2 nm)/Ir20Mn80(10 nm)/Co90Fe10(3 nm)/Cu(2.2 nm)/Co84Fe10B4(10 nm)/Ta(5 nm)] on silicon substrates with 500 nm-thick surface oxide layer was used as a GMR sensor [28, 29]. This sensing element was fabricated using RF magnetron sputtering. The schematic of the SV thin film is illustrated in figure 1(a). The two soft ferromagnetic materials, that is, Cobalt-Iron (CoFe) and Cobalt-Iron-Boron (CoFeB) were employed as pinned- and free-layers, respectively, to realize GMR sensors with large MR and high sensitivity [30]. The CoFe layer served as a pinned layer by pinning from the adjacent antiferromagnetic iridium-manganese (IrMn) layer. Furthermore, owing to its high spin polarization and uniaxial magnetic anisotropy, the CoFeB layer was employed as a free layer, which could be rotated freely following the direction of the applied magnetic field. The non-magnetic copper (Cu) metal layer was selected as a spacer for the two soft ferromagnetic layers because of its high conductivity and interfacial spin-dependent scattering, while Tantalum (Ta) layers served as a buffer and protection layer [30].

A schematic of the home-made measuring system is shown in figure 1(b). The DC in-plane method was used in this system. The Keithley 2401 source meter served as a DC current of 10 mA provider and a voltage output gauge in the Wheatstone bridge circuit. In this setup, the constant-current mode was selected because of its more linear response and higher sensitivity to a small resistance change than the constant-voltage mode because of its small DC current and to protect the sensing system from heating [8]. Furthermore, the Helmholtz coil with a steady power supply was used to generate an in-plane magnetic field parallel to the pinned-layer magnetization direction from 0 to 60 Gauss gradually.

Two types of solutions, Fe3O4 MNPs-ethanol and core–shell Fe3O4@Ag MNPs-ethanol, with equal concentrations (2 mg mL⁻¹), were prepared to verify the Ag coating effect on the properties of MNPs. In addition, to investigate the effect of the number of MNPs on detection performance, a dispersion of Fe3O4 and core–shell Fe3O4@Ag MNPs in ethanol at various concentrations were also prepared. All solutions were sonicated for approximately 15 min to achieve a homogenous solution. Ten microliters of these MNP solutions were drop-cast onto the Ta layer (2 nm) surface of the SV thin film. Also, to measure the voltage change of the SV thin-film, the Cu probes were placed on this surface, which directly contacted the conductive Ta layer. After the ethanol evaporated for approximately 5 min, the real-time output voltage (Vₒ) of the Wheatstone bridge was measured by Keithley 2401 and recorded by the computer for further analysis.
Figure 2. (a) X-ray diffraction profile of the bare Fe₃O₄ (black line) and core-shell Fe₃O₄@Ag (brown line) magnetic nanoparticles with the sign (*), (#), and (o) denote Fe₃O₄, Ag, and α-Fe₂O₃ diffraction peaks. (b) TEM image and Gaussian fitted size distribution of bare Fe₃O₄ and core-shell Fe₃O₄@Ag nanoparticles, and (c), EDS mapping of Fe, Ag, O, and overlap elements.
3. Results and discussion

3.1. Core–shell Fe₃O₄@Ag Nanoparticle Properties

The black precipitate emersion shortly after the stirring of ferrous and ferric salts with the base solution indicates the formation of Fe₃O₄ MNPs through the recognized coprecipitation reaction as follows [31]:

$$\text{Fe}^{2+} + 2\text{Fe}^{3+} + 8\text{OH}^− → \text{Fe}_3\text{O}_4 + 4\text{H}_2\text{O}.$$ 

Figure 2(a) presents the x-ray diffraction profile of both samples, bare Fe₃O₄ (black line) and core–shell Fe₃O₄@Ag (brown line) nanoparticles. The observed diffraction pattern confirmed that Fe₃O₄ is the primary component of the sample by referring to the standard diffraction pattern of Fe₃O₄ (AMCSD 0002400). Another phase of iron oxides, that is, Hematite (α-Fe₂O₃), was also detected as a minor component, manifested by the appearance of only one reflection peak at the (104) plane [32]. By employing the Bragg equation, the calculated lattice spacing of bare Fe₃O₄ MNPs for the most intense diffraction peak observed at 2θ = ~35.5° was ~0.25 nm, which is related to the (311) plane of the Fe₃O₄ crystalline phase [33]. The broadening of the most intense peak (311) was also used to calculate the crystallite size (D) [34] using Debye–Scherer’s equation. The crystallite size of the bare Fe₃O₄ MNPs measured for the (311) plane was ~13 nm, which is in the size range reported by Upadhyay et al [34] for Fe₃O₄ MNPs produced using synthesis temperatures ranging from room temperature to 100 °C. The calculated lattice parameter of 0.8 nm is also in line with another report [34]. In agreement with Iida et al [31], the low intensity of the reflection peaks exhibited by the synthesized Fe₃O₄ sample is a result of the usage of two types of iron salt precursors.

The brown line in figure 2(a) shows the diffraction profile of the core–shell Fe₃O₄@Ag. In addition to the characteristic reflection peaks of Fe₃O₄, reflection peaks at 2θ = 38.15°, 44.34°, and 64.50° with the successive corresponding Miller indices: (111), (200), and (220) reflect the contribution of Ag compared with the standard diffraction pattern of pure Ag (AMCSD 0011135). The Ag coating is probably an amorphous or mixed phase as its crystalline line intensity is very low. Furthermore, considerable impurity is present, which is observed from the numerous unlabeled peaks with low intensity. Owing to the addition of functionalized Ag, the crystallite sizes measured for the (311) plane are just ~12 nm, which is less than that of bare Fe₃O₄. This decrease in crystallite size can be associated with the presence of pores or defects on the Fe₃O₄ surface during functionalization [35].

Furthermore, the particle sizes of both sample bare Fe₃O₄ and core–shell Fe₃O₄@Ag were obtained from the size distribution through the measurement of the diameter from one hundred MNPs for each sample randomly selected from the TEM image. Figure 2(b) shows the TEM image and the associated size distribution of the samples. The bare Fe₃O₄ MNPs and core–shell Fe₃O₄@Ag had a mean diameter of 11 ± 1.8 nm and 17 ± 5.5 nm, respectively. The Gaussian fitting curve revealed that the particle size distribution of core–shell Fe₃O₄@Ag was shifted to a larger size, which confirms the contribution of the Ag coating in the structure. The broadening of the core–shell Fe₃O₄@Ag size distribution observed in figure 2(b) shows the formation of a multiple-core structure (an individual shell particle coats up to several core particles simultaneously) [36] because of the bare Fe₃O₄ agglomeration during the synthesis. Furthermore, as shown in figure 2 (c), the attachment of Ag to Fe₃O₄ was revealed by EDX elemental mapping with Ag composition of ~17%, which approaches the initial raw material composition used to synthesize the Fe₃O₄@Ag MNPs.

Figure 3. Magnetic hysteresis curves comparison of bare Fe₃O₄ and core–shell Fe₃O₄@Ag nanoparticles at room temperature. The left-top inset shows the standard hysteresis curve, while the right-bottom inset shows the enlarged coercive field.
To investigate the effect of Ag coating over the Fe$_3$O$_4$ MNPs surface on its magnetic performance, $M$-$H$ curves of samples were tested using VSM. Figure 3 shows the $M$-$H$ curves of bare Fe$_3$O$_4$ and core–shell Fe$_3$O$_4@Ag$ nanoparticles at room temperature, whereas the inset shows the standard hysteresis loop curve with its magnetic properties. The magnetic properties of the bare Fe$_3$O$_4$ and core–shell Fe$_3$O$_4@Ag$ samples are presented in Table 1. Both samples showed the presence of a hysteresis loop with a low coercive field ($H_C$), which confirms these as magnetically soft nanoparticles. An increase in $H_C$ of Ag-coated Fe$_3$O$_4$ compared to that of bare one is because of the oxidation of MNPs during the core–shell synthesis. Moreover, the Ag coating aggregates particles and increases the effective domain size that can contain at least a weak remanent magnetization. As a consequence of the Ag coating, which induces an increase in the mass of the non-magnetic material and thus reduces the specific magnetization of the whole sample, a notable reduction in the $M_S$ of core–shell NPs was observed, which is in agreement with other reports [15, 21]. Although reduced, the $M_S$ of the Ag-coated Fe$_3$O$_4$ MNPs remained larger than that of the Fe$_3$O$_4$ MNP–graphene magnetic label fabricated by Xu et al, which provided a strong induced-field [2] and was sufficient for particle penetration within the biofilm [21]. The core–shell Fe$_3$O$_4@Ag$ nanoparticle is particularly appropriate as a promising magnetic label for biodetection because it merges two eminences of large induced-field and excellent biocompatibility. This hybrid MNP exhibits appropriate magnetic properties because of the Fe$_3$O$_4$ core presence and anti-bacterial activity as a result of the Ag surface modification [21], which broadens its bio-functionalities beyond their individual single elements.

### 3.2. Effect of silver coating on Fe$_3$O$_4$ nanoparticles

Figure 4 shows the magnetic field dependence of the output voltage for the SV thin-film, SV thin-film with Fe$_3$O$_4$ MNPs, and SV thin-film with core–shell Fe$_3$O$_4@Ag$ MNPs. This dependence can be explained using the spin-dependent transport principle. Initially, in the absence of a magnetic field ($H = 0$), $V_{O}$ was equal to zero. As the magnetic field was magnified, the voltage drastically increased until it was saturated at ~12 Gauss. This step-like $V_{O}$ change is the advantage of this sensor, which can be solved effortlessly without further signal processing [38]. As the MNPs are placed on the SV thin-film surface, the magnetic switching field enlarges by ~6 Gauss, indicating MNP detection.

| Nanoparticle samples | $M_S$ (emu/g) | $H_C$ (Oe) |
|----------------------|--------------|------------|
| Fe$_3$O$_4$          | 77           | 51         |
| core–shell Fe$_3$O$_4@Ag$ | 53          | 145        |

Figure 4. Magnetic field dependence of the output voltage.
The different levels of saturated $V_O$ in the bare SV thin-film compared with the MNPs present in figure 4 are a result of the different magnetization magnitude of the free layer, as illustrated in figure 5. Moreover, as the Fe$_3$O$_4$ MNPs are bound on the SV thin-film, their magnetic moment ordering plays an important role because of the individual induced-field ($h$) generated by each MNP [1, 7, 8, 39]. The ordered magnetic moments generate a non-zero net induced-field ($h_{\text{net}} = 0$) that reduces the effective magnetic field exposed to the free layer [8] of the SV thin film. This causes a decline in the conductivity and saturation of $V_O$. Meanwhile, as the core–shell Fe$_3$O$_4$@Ag MNPs are attached to the SV thin film, the saturated $V_O$ magnifies because of the $M_S$ drop of core–shell Fe$_3$O$_4$@Ag MNPs.

As shown in figure 4, the external magnetic field at the saturated $V_O$ of the integrated SV-MNP system is reduced (to a few Gauss) compared to that at the saturated magnetization of the MNPs, which is in the range of kOe (figure 3). This is because of the magnetic interaction between the non-linear net magnetization of the MNPs and the free layer of the SV thin-film, which stimulates the change in their individual magnetization behaviors [40]. The saturated $V_O$ of the SV thin-film, which is almost twice as high as that with the integrated SV-MNP system, can be attributed to the generated induced-field by MNPs.
3.3. Detection of core–shell Fe₃O₄@Ag nanoparticles with various concentrations

To observe the effect of the core–shell Fe₃O₄@Ag MNPs number on the detected signals, the $V_O$ was measured for various MNP solution concentrations. Higher MNP concentrations present larger numbers of MNPs. $V_O$ of the SV thin-film with only 10 μL of ethanol (without core–shell Fe₃O₄@Ag MNPs) was used as a control. Figure 6 shows the $V_O$ upon an applied magnetic field for various core–shell Fe₃O₄@Ag nanoparticle concentrations. $V_O$ starts to saturate immediately after exposure to a certain magnitude of magnetic field. The magnitude of saturated $V_O$ is significantly affected by the concentration of MNPs. The increasing number of core–shell MNPs reduces the magnitude of the saturated $V_O$ through the reinforcement of the MNP induced-field.

The effect of MNPs of various concentrations on the GMR sensor signal ($ΔV$) is presented in figure 7, where the dashed lines represent the fitted curve. The signal is measured by taking the average of the saturated $V_O$ after being corrected with the background, while the error bar is obtained from its standard deviation. In contrast, $V_O$ of the bare SV thin-film is performed as a background. The signal increased linearly with the addition of the core–shell Fe₃O₄@Ag MNPs, which is in agreement with another result [7]. This linear response manifests the MNPs detection and accordingly, it can be employed as a specific bio-assay method through binding to particular antibodies. An increase in $ΔV$ originates from the magnification of the induced-field from the MNPs. Considering the measured core–shell Fe₃O₄@Ag MNPs concentration, the GMR sensor possesses two different sensitivities obtained from the gradient of the linear fitted curves. For high concentrations of core–shell MNPs (>2 g L⁻¹), the sensor sensitivity was 0.02 mV g⁻¹ L⁻¹. This sensor is more sensitive to low concentration (<2 g L⁻¹) detection with 0.76 sensitivity of sensitivity, even though it is still lower than the sensitivities reported in other studies that used commercial SV-based sensors in detecting iron-based MNPs [2, 7, 8, 39]. The lower sensitivity of high core–shell MNPs concentrations can be attributed to the magnetic responses of an MNP ensemble. High core–shell MNP concentrations lead to a decrease in the inter-particle distances and accordingly, the dipole–dipole interaction induced by the adjacent MNPs is reinforced [41]. This stronger dipole interaction reduces the effective magnetic field acting on each MNP, which subsequently results in a weaker magnetic response [41].

Furthermore, as shown in figure 7, the sensor signal dependence of the MNPs concentration from 0.2 g L⁻¹ to 50 g L⁻¹ for Fe₃O₄ exhibiting nonlinear relations with a negative exponential function fits well. For higher concentrations (>50 g L⁻¹), the sensor no longer responds to an increase in MNP concentration, which is represented by two out-fit data points. The weak response of the sensor for high Fe₃O₄ MNPs concentrations can be attributed to the stronger dipole interactions of the adjacent Fe₃O₄ MNPs for unprotected surfaces, which leads to a very weak magnetic response.

Figure 7. Magnetic nanoparticles concentration dependence of the GMR sensor signal.
Based on this discussion, further research is needed to examine the effectiveness of the present sensor system and compare it with the commercial SV-based GMR sensor. A comparative study of ferromagnetic NPs of Fe3O4 with commercial super-paramagnetic Fe3O4 NPs is required. Similarly, a comparative study of the effect of the concentration of core–shell Fe3O4 (ferromagnetic) @Ag MNPs with core–shell Fe3O4 (super-paramagnetic) @Ag MNPs on signal detection is required to acquire a better core–shell magnetic label.

4. Conclusions

A biocompatible core–shell Fe3O4@Ag MNPs magnetic label with sizable saturation magnetization was synthesized to analyze the performance of a spin-valve thin-film based GMR sensor with the Wheatstone bridge. It was revealed that the sensor successfully distinguished between Fe3O4@Ag and Fe3O4@Ag MNPs through an increase in the output voltage. The output signal of the sensor responds linearly to an increase in the number of core–shell Fe3O4@Ag magnetic nanoparticles. This sensor performs better on core–shell Fe3O4@Ag nanoparticle detection at low concentrations. These results suggest that the combination of biocompatible core–shell Fe3O4@Ag nanoparticle magnetic labels with a spin-valve thin-film based GMR sensor is extremely promising for utilization in real magnetic-based bio-detection.

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