Fabrication of New Fe₃O₄/PVA/(C₆H₇O₆Na)n Nanohybrid Ferrogels for Antibacterial Applications

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New Fe₃O₄/polyvinyl alcohol (PVA)/sodium alginate (C₆H₇O₆Na)n nanohybrid ferrogels for antibacterial applications were fabricated. The crystal and molecular structures along with optical and magnetic properties of the prepared samples were characterized. The antibacterial activity of the ferrogels against Bacillus subtilis and Escherichia coli was investigated using the agar dilution method. X-ray diffraction analysis showed that the Fe₃O₄/PVA comprised a PVA amorphous phase and a spinel-structured Fe₃O₄ crystalline phase. The Fe₃O₄/PVA crystallite size was 7.5–9.9 nm and the scanning electron micrographs showed that the Fe₃O₄/PVA agglomerated. The ferrogels were superparamagnetic with saturation magnetizations from 14.8 × 10⁻³ to 82.1 × 10⁻³ emu/g. The absorption of the ferrogels showed a bathochromic effect, accompanied by an increase in the bandgap from 2.09 to 2.18 eV with increasing Fe₃O₄ content in the ferrogels. The ferrogels demonstrated new potency as antibacterial agents against B. subtilis and E. coli, where their antibacterial performance increased with increasing Fe₃O₄ content.

Keywords: Fe₃O₄/PVA/(C₆H₇O₆Na)n, nanohybrid, ferrogels, superparamagnetic, antibacterial agent.

1. Introduction

Over the past few decades, researchers have widely developed methods of preparing Fe₃O₄ nanoparticles (NPs) in various shapes and sizes and in the forms of powders, films, ferrofluids, and ferrogels. In particular, intensive research on Fe₃O₄ NPs in the form of ferrogels has been motivated by their advantageous properties compared with those of other forms, even ferrofluids. Theoretically, Fe₃O₄ ferrogels, which are gels with a cross-linked polymer network containing Fe₃O₄ NPs as a filler, offer specific advantages over conventional gels with respect to their high flexibility and high sensitivity to an external magnetic field, which implies that ferrogels can be easily positioned inside a living organism1.

Fe₃O₄ NPs exhibit superior advantageous properties, including superparamagnetism, biocompatibility, and low toxicity. They are also inexpensive and easily synthesized2, making such materials attractive in the biomedical field3, for example, as antibacterial agents for skin infections. In general, skin infections occur and often pose therapeutic challenges because of increasing concerns about multidrug-resistant bacteria4. Furthermore, gel-based skin antibacterial agents are particularly attractive because gels have advantages over powders and liquids. However, the Fe₃O₄-based antibacterial agents developed thus far have been based on powders. Therefore, the development of Fe₃O₄ ferrogels as antibacterial agents is strongly desired.

Fe₃O₄ ferrogels based on polyvinyl alcohol (PVA) have recently begun to attract researchers’ attention for possible use in various multipurpose applications5–7. Recent research has shown that Fe₃O₄ ferrogels with a PVA matrix have excellent prospects for use in biomedical applications8. However, for skin antibacterial applications, PVA has a drawback because of its relatively high stiffness, which reduces antibacterial efficacy. Consequently, an appropriate biocompatible crosslinking agent that reduces the stiffness of Fe₃O₄/PVA ferrogels is urgently needed.

Sodium alginate, with chemical formula (C₆H₇O₆Na)n, exhibits strong potential for use as a crosslinking agent that supports the antimicrobial performance of Fe₃O₄/PVA ferrogels. Theoretically, in addition to reducing the stiffness of PVA5, (C₆H₇O₆Na)n offers other advantages of nontoxicity, biocompatibility9, biodegradation ability, good mechanical properties, and relatively low cost10,11. Therefore, in the present work, we developed new Fe₃O₄/PVA/(C₆H₇O₆Na)n nanohybrid ferrogels as antibacterial agents and investigated their crystal and molecular structures, optical properties, bandgaps, magnetic behaviors, and antibacterial efficacy.
2. Experimental Methods

2.1 Materials

The main precursors were FeCl$_2$·6H$_2$O, FeCl$_3$·6H$_2$O, HCl (12 M), NH$_4$OH (6.5 M), PVA (M$_w$ = 60,000), and (C$_6$H$_7$O$_6$Na)$_n$. All precursors were analytical grade to ensure high purity and quality of the samples. All precursors were purchased from Merck and were used without further purification.

2.2 Fabrication of Fe$_3$O$_4$/PVA Nanocomposites

The Fe$_3$O$_4$ NPs were prepared using a coprecipitation method following the procedure described in our previous work$^{13}$. One g of PVA was dissolved in 20 mL of distilled water at 120 °C for 3 h using a magnetic stirrer with a speed of 720 rpm. Various masses of Fe$_3$O$_4$ (0.13, 0.26, 0.39, 0.52, and 0.65 g) were mixed with 2 mL of PVA solution at 120 °C for 30 min using a magnetic stirrer with a speed of 720 rpm to produce Fe$_3$O$_4$/PVA. The Fe$_3$O$_4$/PVA was collected after the mixture cooled to room temperature. The samples were labeled S1–S5 according to the respective masses of Fe$_3$O$_4$ (0.13, 0.26, 0.39, 0.52, and 0.65 g).

2.3 Fabrication of Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ Ferrogels

The Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels were prepared through a sonochemical method. Three hundred mg of (C$_6$H$_7$O$_6$Na)$_n$ was dissolved in 20 mL of distilled water at 120 °C under stirring using a magnetic stirrer with a speed of 720 rpm for 4 h, followed by a cooling process to room temperature. Next, 2 mL of (C$_6$H$_7$O$_6$Na)$_n$ solution was reacted with the prepared Fe$_3$O$_4$/PVA (S1–S5 samples) at 120 °C for 30 min under stirring using a magnetic stirrer with a speed of 720 rpm. After the mixtures cooled to room temperature, the final products were obtained as solid-black gels in the form of Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels. The prepared Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels were labeled SF1–SF5 according to the respective S1–S5 samples. The schematic showing the construction of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels is shown in Figure 1.

2.4 Characterization

The structural, optical, and magnetic properties were characterized by X-ray diffraction (XRD) for S1–S5 samples, scanning electron microscopy/energy-dispersive X-ray spectroscopy (SEM/EDX) for S1–S5 samples, UV–Vis spectrophotometry for SF1–SF5 samples, Fourier transform infrared (FTIR) spectroscopy for SF1–SF5 samples, and vibrating sample magnetometry for SF1–SF5 samples. The antimicrobial performance of the SF1–SF5 samples against *Bacillus subtilis* (Gram-positive) and *Escherichia coli* (Gram-negative) was evaluated through the agar diffusion method.

All characterizations were conducted at ambient temperature.

3. Results and Discussion

The XRD patterns of the Fe$_3$O$_4$/PVA used as the filler of the ferrogels (Figure 2) show that two phases were formed, i.e., amorphous and crystalline phases. The amorphous phase, with a broad diffraction peak detected at 2θ of approximately 19.5°, was attributed to PVA. This is consistent with the results of the previous work$^{14}$. Meanwhile, the XRD patterns of the crystalline phase showed diffraction peaks at 2θ of 30.1°, 35.6°, 43.3°, 57.3°, and 63.0°, which represent the (200), (311), (400), (422), (511), and (440) planes of Fe$_3$O$_4$, respectively$^{15}$. Furthermore, the intensities of the Fe$_3$O$_4$ peaks were observed to increase with increasing Fe$_3$O$_4$ content. Interestingly, the increase in Fe$_3$O$_4$ content led to a decrease in the intensity of the peak of the PVA amorphous phase because of the reduced PVA content. The crystallinity, i.e., the degree of regularity and structural order in the Fe$_3$O$_4$/PVA

Figure 1. Schematic showing the construction of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels.
system, was found to increase from 22.8% to 61.0% with increasing Fe$_3$O$_4$ content.

The Fe$_3$O$_4$ crystal structure in this study exhibited a cubic spinel structure. The lattice parameters of the Fe$_3$O$_4$ in all samples were constant, with a value of $a = b = c = 8.365$ Å.

In this crystal structure, the Fe$^{2+}$ and Fe$^{3+}$ ions are distributed in octahedral and tetrahedral sites. The distribution of these metallic ions determines the magnetic moments of Fe$_3$O$_4$ where the magnetic moments between ions in octahedral and tetrahedral sites are anti-parallel. Furthermore, the crystallite size ($D$) of the Fe$_3$O$_4$ of all samples was calculated using the Debye–Scherrer equation: $D = \frac{0.9\lambda}{\beta\cos\theta}$, where $\lambda$, $\beta$, and $\theta$ are the wavelength of the X-rays, the full-width at half-maximum of the peak, and the diffraction angle, respectively$^{16}$. The data analysis results in Table 1 show that $D$ increased with increasing Fe$_3$O$_4$ content. The increasing Fe$_3$O$_4$ content tended to increase the extent of aggregation into clusters because of the concomitant increase in the

![Figure 2. XRD patterns of the Fe$_3$O$_4$/PVA.](image)

![Figure 3. SEM images of the Fe$_3$O$_4$/PVA.](image)

| Sample | $a = b = c$ (Å) | Crystallite size (nm) | Crystallinity (%) |
|--------|-----------------|-----------------------|-------------------|
| S1     | 8.365           | 7.5                   | 22.8              |
| S2     | 8.365           | 8.0                   | 30.6              |
| S3     | 8.365           | 8.9                   | 46.2              |
| S4     | 8.365           | 9.4                   | 53.4              |
| S5     | 8.365           | 9.9                   | 61.0              |

Table 1. Crystallite parameters of the Fe$_3$O$_4$/PVA
van der Waals force. This result is consistent with the SEM images, as shown in Figure 3. The SEM images show that the Fe$_3$O$_4$ was covered with PVA, which led to aggregation. As the Fe$_3$O$_4$ content was increased, the Fe$_3$O$_4$/PVA tended to form more clusters because of the reduced PVA content. Therefore, the Fe$_3$O$_4$ NPs, which have a high specific-surface-area-to-volume ratio, tended to agglomerate because of the interaction of magnetic particles or because of van der Waals force. In this regard, Roohani et al. showed that aggregation occurs as a result of hydrogen bonds between particles because PVA, which is rich in hydroxyl groups, cannot cover all of the hydrogen bonding sites of the Fe$_3$O$_4$ NPs. The elemental composition of the samples is presented in Table 2. The elemental analysis results indicate that the Fe content increases and the C content decreases with increasing Fe$_3$O$_4$ used in the synthesis.

The transmittance spectra used to characterize the functional groups of the Fe$_3$O$_4$/PVA/(C$_6$H$_5$O$_n$Na) ferrogels are presented in Figure 4. The Fe–O stretching band originating from Fe$_3$O$_4$ is observed at 420–500 cm$^{-1}$ for the tetragonal sites and at 700 cm$^{-1}$ for the octahedral sites. Furthermore, stretching bands characteristic of PVA were detected at 857 cm$^{-1}$ (CC stretching)$^{19}$, 1047–1147 cm$^{-1}$ (C–O)$^{20}$, 1350 cm$^{-1}$ (C–O–C), and 2966 cm$^{-1}$ (CH$_2$ asymmetric stretching)$^{21}$. The stretching bands characteristic of (C$_6$H$_5$O$_n$Na) were detected in the range 1473–1560 cm$^{-1}$ (C=O asymmetric stretching) and at 1708 cm$^{-1}$ (C=O). Interestingly, the Fe$_3$O$_4$, PVA, and (C$_6$H$_5$O$_n$Na)$_n$ as the main components of the ferrogels, all contributed to the broad O–H band between 3305 and 3670 cm$^{-1}$.

The contribution of Fe$_3$O$_4$, PVA, and (C$_6$H$_5$O$_n$Na) to the functional bonding observed by the FTIR spectra initiates further discussion. The changes in intensities of chemical bands provide a clue on the mechanical reorientation of the molecules.$^{24}$ The decreasing intensities of O–H band, CH$_2$ asymmetric stretching, =C–O–C, and C–O bonding is believed due to the decrease of PVA content. This trend is related to the increase of Fe$_3$O$_4$ and the presence of alginate. From the theoretical viewpoint, the presence of alginate reduces the stiffness of PVA-based system by virtue of its low elongation break.$^{25}$ In other words, the long-chain structure of PVA in Fe$_3$O$_4$/PVA nanocomposites was destroyed by the addition of alginate. Zhang et al. claimed that one of the best ways to reduce the stiffness of fabric-reinforced PVA was by introducing Na-alginate.$^{26}$ They discovered that mechanical strength and stiffness could be decreased due to the swelling behavior of composite hydrogels that diminish friction amongst interfibers.$^{26}$ Furthermore, alginate is not only effective in reducing the stiffness of PVA-fibers, but also other polymers, such as poly-lactic-co-glycolic acid.$^{27}$

The absorbance spectra, as shown in Figure 5, were recorded to investigate the optical properties of the Fe$_3$O$_4$/PVA/(C$_6$H$_5$O$_n$Na) ferrogels. The peaks at 474 and 511 nm are consistent with the previous work.$^{28}$ The absorbance intensity is associated with the number of particles present. Meanwhile, the wavelength of maximum absorption is associated with the particle size. Physically, these peaks represent the difference in energy levels between the $\sigma$ and $\pi^*$ states.$^{29}$ In addition, incorporating the Fe$_3$O$_4$ into ferrogels would increase the distribution of particles embedded in the (C$_6$H$_5$O$_n$Na) crosslinking agent. As a consequence, the conduction electrons on the surface of the Fe$_3$O$_4$ migrate from one atom to neighboring atoms so that the surface plasmon resonance shifts toward lower energies, which is associated with smaller wavelengths.$^{30}$ In physics, this phenomenon is described as the bathochromic effect. The bandgap ($E_g$) of the Fe$_3$O$_4$/PVA/(C$_6$H$_5$O$_n$Na) was calculated using the equation $\alpha h \nu = A(h \nu - E_g)^{1/2}$, where $\alpha$, $h$, $\nu$, and $A$ are the absorption coefficient, Planck’s constant, the frequency, and a constant, respectively.$^{31}$ The linear part of the curve was extrapolated to the intersection with the energy axis to determine the bandgap.$^{32}$ The data analysis is shown in Figure 6. The $E_g$ of the Fe$_3$O$_4$/PVA/(C$_6$H$_5$O$_n$Na) ferrogels.

### Table 2. Elemental composition of the Fe$_3$O$_4$/PVA

| Sample | Wt. (%) |
|--------|---------|
| Fe     | O       | C       |
| S1     | 9.6     | 35.7    | 54.7    |
| S2     | 12.9    | 37.1    | 50.0    |
| S3     | 22.7    | 35.3    | 42.0    |
| S4     | 29.9    | 30.5    | 39.6    |
| S5     | 34.2    | 30.9    | 34.9    |

**Figure 4.** FTIR spectra of the Fe$_3$O$_4$/PVA/(C$_6$H$_5$O$_n$Na) ferrogels.

**Figure 5.** Absorbance spectra of the Fe$_3$O$_4$/PVA/(C$_6$H$_5$O$_n$Na) ferrogels.
increased from 2.09 eV to 2.18 eV with increasing Fe$_3$O$_4$ content. This phenomenon is explained by Fe$_3$O$_4$ reduced dissociation as a result of its increased dispersion in the ferrogels. It implies that collisions among electrons in the magnetic particles were reduced, resulting in an increase of the energy on the surface. Other authors have reported an $E_g$ of slightly greater than 2.2 eV for Fe$_3$O$_4$\(^{33}\). This difference is attributed to the effect of the surface modification of Fe$_3$O$_4$ using PVA, which led to dispersion of the Fe$_3$O$_4$ in (C$_6$H$_7$O$_6$Na)$_n$ to form nanohybrid ferrogels.

The magnetization curves of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels are depicted in Figure 7. The figure shows that all of the samples are likely to exhibit superparamagnetic character, as indicated by the characteristic S-like shape with the coercivity field close to zero\(^{34}\). A quantitative analysis was carried out to fit the magnetization curves using the Langevin equation:

$$M = M_s \coth \left( \frac{\mu H}{k_B T} \right) - \left( \frac{k_B T}{\mu H} \right)$$

where $M$, $M_s$, $\mu$, $H$, $k_B$, and $T$ are the magnetization, saturation magnetization, magnetic moment, Boltzmann constant, and temperature, respectively. The magnetic parameters obtained from the data analysis are presented in Table 3. The magnetic moment of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels increased by the addition of Fe$_3$O$_4$ content. The magnetic moment plays an essential role in a material’s response to an external magnetic field. Interestingly, not only the increase in Fe$_3$O$_4$ content but also the increase in particle size contributed to

**Table 3. Magnetic parameters of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels**

| Sample | $M_s$ ($\times 10^{-3}$ emu/g) | $H_c$ (T) | Magnetic moment ($\times 10^{-19}$ J/T) |
|--------|-------------------------------|-----------|--------------------------------------|
| SF1    | 14.8                          | 0.013     | 8.1                                  |
| SF2    | 16.8                          | 0.016     | 8.7                                  |
| SF3    | 42.1                          | 0.020     | 9.5                                  |
| SF4    | 64.3                          | 0.025     | 10.1                                 |
| SF5    | 82.1                          | 0.028     | 14.0                                 |

Figure 6. Bandgaps of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels.
the increase in $M_s$. Karaagac et al. have similarly reported that particle size strongly influences the $M_s$\textsuperscript{35}. Furthermore, the coercivity field also increased with increasing Fe$_3$O$_4$ content, which is attributable to the accompanying increase in particle size. Previous work has shown the same trend for Fe$_3$O$_4$/PVA with a ribbon-like structure\textsuperscript{36}.

The antibacterial performance of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$, ferrogels against *B. subtilis* and *E. coli* is presented in Figure 8. This figure shows that the inhibition-zone diameter increases with increasing Fe$_3$O$_4$ content. The inhibition-zone is associated with the antibacterial performance of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels. Theoretically, the ability to destroy bacteria is strongly correlated with the ability of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ to generate reactive oxygen species (ROS)\textsuperscript{37}. ROS generate superoxide as a free radical that inhibits bacterial growth\textsuperscript{38}. Furthermore, in the case of Fe$_3$O$_4$ NPs incorporated into ferrogels, the inhibition-zone diameter was larger than that of pure Fe$_3$O$_4$. The enhanced bactericidal activity of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels is attributed to the better dispersion of the Fe$_3$O$_4$ particles in the ferrogels than in the pure Fe$_3$O$_4$, which tends to aggregate. In addition to ROS, particle size also plays an important role in bactericidal activity. Other authors have shown that Fe$_3$O$_4$ with small particle size and monodisperse shape exhibits better inhibitory activity against bacteria than Fe$_3$O$_4$ with large particle size and polydisperse shape\textsuperscript{39}. Theoretically, Fe$_3$O$_4$ NPs with an excellent particle size distribution are likely to penetrate more easily into the walls or pores of bacteria. As a comparison, Jalali et al. prepared Fe$_3$O$_4$ NPs by doping Co and Zn to strengthen their antibacterial potential; however, they observed an inhibition-zone diameter of only 6.4 mm for *E. coli*\textsuperscript{40}. In line with that, Salem and co-workers fabricated Fe$_3$O$_4$ NPs from marine algae; however, the inhibition zone diameter for *E. coli* was still approximately 6 mm and that for *B. subtilis* was approximately 5 mm. These values are still inferior compared with that of the ferrogels investigated in the present study.

The bactericidal mechanism of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels is shown in Figure 9. Fe$_3$O$_4$ NPs generate reactive oxygen species (ROS), which destroy the microbe’s nuclei and DNA.
The bactericidal mechanism of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels is presented in Figure 9. From a physics perspective, the negative surface charges on the Fe$_3$O$_4$/PVA nanocomposite particles repel the bacteria under electrostatic interaction mechanism. This repulsion makes the metal oxide NPs less effective in destroying bacteria. However, when Fe$_3$O$_4$/PVA are dispersed in (C$_6$H$_7$O$_6$Na)$_n$, the net charge at the surface of the particles becomes positive. This change in surface charge is beneficial in terms of catching the bacteria by Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels. Thus, the chance of ROS penetrating into the bacteria increases. This leads to increase in the diameter of the inhibition zone. Thus, the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels exhibit superior antibacterial activity. Furthermore, because of the structural complexity of the Fe$_3$O$_4$ ferrogels, a theoretical explanation of the performance of the ferrogels related to their magnetic-sensitive behavior has not yet been established. Therefore, the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels to be used on a large scale, further investigation is necessary, especially with respect to the optimum filler, matrix, cross-linker contents in the ferrogels.

4. Conclusion

We synthesized new Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels using a combination of coprecipitation and sonochemical methods. The XRD data indicated the presence of two phases: amorphous PVA and crystalline Fe$_3$O$_4$. The crystallite size and crystallinity increased with the addition of Fe$_3$O$_4$. The surface of the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ agglomerated because of the increase in Fe$_3$O$_4$ content, preventing PVA from covering it optimally. The nanohybrid ferrogels exhibited superparamagnetism, where the coercivity of the particles becomes positive. This change in surface charge is beneficial in terms of catching the bacteria by Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels. Thus, the chance of ROS penetrating into the bacteria increases. This leads to increase in the diameter of the inhibition zone. Thus, the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ nanohybrid ferrogels exhibit superior antibacterial activity. Furthermore, because of the structural complexity of the Fe$_3$O$_4$ ferrogels, a theoretical explanation of the performance of the ferrogels related to their magnetic-sensitive behavior has not yet been established. Therefore, the Fe$_3$O$_4$/PVA/(C$_6$H$_7$O$_6$Na)$_n$ ferrogels to be used on a large scale, further investigation is necessary, especially with respect to the optimum filler, matrix, cross-linker contents in the ferrogels.

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