Facile fabrication of green synthesized silver-decorated magnetic particles for coating of bioactive packaging

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Abstract To avoid bacterial and viral infections on food products, the use of antibacterial and antiviral packaging offers great benefit to the food industry. In this study, the coating of paper packaging with silver-decorated magnetic particles (Ag@Fe₃O₄) was developed. The Ag@Fe₃O₄ was prepared by a facile and environmentally friendly method using extracted spent coffee grounds (ex-SCG). The effects of Ag@Fe₃O₄ content on properties of coated paper were investigated. The overall properties of coated paper improved when the Ag@Fe₃O₄ content increased up to 0.15%w/v. An increase in tensile strength of 154.01% and a decrease in water vapor permeability of 48.50% were found in coated paper with 0.15%w/v Ag@Fe₃O₄. Furthermore, the coated paper also exhibited the synergistic effect on antibacterial activities against Escherichia coli (E. coli) and Staphylococcus aureus (S. aureus). The release of metal ions in food simulants and kinetic release parameters were also studied. The release of silver ions and ferrous ions in food simulants met the requirement of overall migration limit of the European Standard. The paper coated with 0.15%w/v Ag@Fe₃O₄ had better capabilities to maintain quality and extend shelf-life of tomatoes. The obtained Ag@Fe₃O₄ coated paper is promising for bioactive food packaging to retain food freshness.

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Introduction

Due to the current COVID-19 pandemic, the use of antibacterial and antiviral materials has attracted much attention to avoid infections by bacteria and viruses (Imani et al. 2020). Many kinds of materials have been developed to have the properties integrated with an antibacterial function (Al-Tayyar et al. 2020). Nowadays, food packaging plays an important role in contributing to our everyday lives and many industries, especially horticulture production. The packaging can improve shelf life, minimize postharvest loss and ensure product quality while transportation (Alhendi and Choudhary 2013; Verma et al. 2021; Fernandez et al. 2022). The current trend of food packaging development also focuses on antibacterial packaging that can extend the shelf-life and maintain the quality of food products (Chawla et al. 2020). Ounkaew et al. (2022) observed that the bioactive films containing zinc oxide and citric acid could inhibit the bacterial growth in cherry tomatoes during the period of storage and delay the spoilage, thus prolonging the shelf-life of products.

Cellulosic paper is extensively used as packaging to protect foodstuff from external damage and contamination due to its recyclability, biodegradability, and low price (Shankar et al. 2019; de Oliveira et al. 2022; Zhang et al. 2020). The coating of paper to enhance its properties is a common practice (Rastogi and Samyn 2015; Mortazavi et al. 2021). The coated paper with bioactive agents is an option to achieve active food packaging (Apjok et al. 2019). Battisti et al. (2017) prepared the biopolymeric coating based on gelatin solution containing antibacterial and antioxidant agents such as citric acid for beef packaging. The presence of 0.5–1% w/w of citric acid in the coated paper could decrease the spoiling and rotting rates of beef. Among the biopolymers used to prepare food packaging, carboxymethyl cellulose (CMC) has been widely used owing to its biodegradability, non-toxicity and low cost (Atta et al. 2021a, b). CMC can be blended with a biocompatible polymer such as polyvinyl alcohol (PVA) to enhance the physical and mechanical properties (Siregar et al. 2019). Muppalla et al. (2014) found that the incorporation of 4% citric acid into PVA/CMC/aloe vera film led to the formation of ester bonds resulting in 75% increase in tensile strength.

Reinforcement of biopolymers with metal particles is another approach to improve mechanical properties, water resistance and antibacterial properties (Kumar et al. 2021). Magnetic particles (Fe₃O₄) have been considered as a good candidate for reinforcing biopolymers in food packaging applications because of their biocompatibility, non-toxicity, and low price with antibacterial and antioxidant activities. Recently, the combination of Fe₃O₄ with silver nanoparticles (Ag@Fe₃O₄) has been developed as an antimicrobial agent in various medical, biological, food packaging, and textile applications (Hatami et al. 2019; Keshk et al. 2018). Saedi and Rhim (2020) prepared active packaging film containing Fe₃O₄ and Ag@Fe₃O₄. The presence of Ag@Fe₃O₄ produced high antibacterial activity over 12 h testing. The intensity of silver ions on Fe₃O₄ surface could assist the inhibition of bacterial growth. Ag@Fe₃O₄ can be synthesized via thermal decomposition, microemulsion, and green synthesis. For the green synthesis of Ag@Fe₃O₄, Ghaseminezhad and Shojaosadati (2016) synthesized Ag@Fe₃O₄ for the first time using Fe₃O₄ surrounded with hydroxyl groups of starch as bio-reducing agent to reduce silver ions. Dehghan et al. (2022) reported green synthesis of Ag@Fe₃O₄ using leaf extract of Eryngium planum. The Ag@Fe₃O₄ can inhibit S. aureus, E. coli as well as Cryptococcus neoformans.

Currently, there is a dearth of information on green synthesis of Ag@Fe₃O₄ by food and beverage wastes and their applications. Hence, this study aimed to develop a novel packaging paper coated with Ag@Fe₃O₄. The CMC/PVA crosslinked with citric acid
was applied as polymer matrix. For the synthesis of Ag@Fe₃O₄, the ex-SCG was used as a bio-reducing agent for the green synthesis of Ag@Fe₃O₄. The effect of Ag@Fe₃O₄ at 0–0.2 wt% on properties of coated paper viz., mechanical properties, antibacterial activities and water resistance were investigated. In addition, the coated paper was used as packaging for tomatoes. The qualities of the tomatoes in such packaging were also studied.

**Experiment**

**Materials**

CMC (99.99% purity) and PVA (Mw of 1700–1800) were supplied from Laboratory reagents & Fine Chemical, Thailand. Silver nitrate (AgNO₃), and citric acid and sodium hydroxide were obtained from RCI Labscan Limited. Spent coffee grounds (SCG) in term of powder were collected from Vector coffee roster, Khon Kaen, Thailand. Glycerol, sodium hydroxide (NaOH), magnesium chloride (MgCl₂), sodium chloride (NaCl), magnesium nitrate Mg(NO₃) and potassium carbonate (K₂CO₃) were purchased from KEMAUS. Iron (III) chloride hexahydrate (FeCl₃·6H₂O), and Iron (II) sulfate heptahydrate (FeSO₄·7H₂O) were from AppliChem and KEMAUS, respectively. Filter paper (whatman, No. 1) was used as a substrate for coating.

**Preparation of Fe₃O₄**

Four grams of FeCl₃·6H₂O was mixed with 125 mL deionized water and stirred at 550 rpm at 90 °C for 10 min. Then 2.7 g of FeSO₄·7H₂O was added into the mixture and stirred for another 30 min. NaOH solution was prepared by dissolving 10 g of NaOH in 40 mL deionized water. The NaOH solution was added into as prepared solution and the Fe₃O₄ was then separated using magnet and oven-dried at 40 °C for 5 h. The particle was then rinsed with DI water until pH of 7.0 was obtained and oven-dried at 40 °C for another 5 h.

**Microwave assisted synthesis of Ag@Fe₃O₄**

The ratio of SCG to deionized water was fixed at 1:10 in this case. The SCG was mixed with deionized water under mechanical stirring at 90 °C for 5 min. Then, the solution was filtered through qualitative filter and and centrifuged at 3500 rpm for 5 min to obtain the ex-SCG with total phenolic content of 6.68 mg/L gallic acid equivalent. Total phenolic content was determined using Folin Ciocalteu method according to Trongchuen et al. (2018) with minor modification. The ex-SCG was mixed with Fe₃O₄ and 100 mM of AgNO₃. The solution was mechanically stirred for 2 min and poured into a glass container with a lid. The reaction took place in a microwave oven (R-219, SHARP) for 30 min. The Ag@Fe₃O₄ was separated using magnet and oven-dried at 40 °C for 5 h. The element component of Ag@Fe₃O₄ was analyzed using X-ray fluorescence (S8 TIGER XRF spectrometer, Bruker AXS). The ratio of Ag:Fe₃O₄ was 1:22.

**Preparation of paper coated with Ag@Fe₃O₄**

One gram of CMC was dissolved in 25 mL of ex-SCG. One gram of PVA in 25 mL of ex-SCG was prepared at 90 °C and left to cool down at room temperature. The CMC and PVA solutions were mixed and then added with 0.05, 0.1, 0.15 and 0.2% w/v of Ag@Fe₃O₄ using ultrasonication for 30 min to disperse Ag@Fe₃O₄. Half a milliliter of glycerol and 0.75 g of citric acid were mixed and stirred for 5 min. The composite solution with 50 mL was cast on the paper with area of 150 cm² using a Doctor blade with a gap of 200 µm.

**Characterization of paper coated with Ag@Fe₃O₄**

The Attenuated total reflectance Fourier Transform Infrared Spectroscopy (ATR-FTIR) of 2 cm × 2 cm paper coated with Ag@Fe₃O₄ was carried out using FTIR, Bruker TENSOR27. The spectrum was analyzed at 4000–600 cm⁻¹ with at a resolution of 2 cm⁻¹ for 64 scans.

The X-ray diffraction (XRD) of paper coated with Ag@Fe₃O₄ was tested by a SmartLab X-ray diffractometer provided with a Cu Kα radiation source operated at 45 kV and 200 mA. A paper coated with Ag@Fe₃O₄ with dimensions of approximately 2 cm × 2 cm was placed on the exposing stage and scanned at diffraction angles of 2θ = 10–70°.

Samples with dimensions of 25 mm × 80 mm × 0.3 mm were prepared for
measurement of tensile strength using universal testing machine, UTM (Instron, Model 5567). The gauge length was set at 50 mm and a crosshead speed of 25 mm min\(^{-1}\) were used in accordance with ASTM D 882–10. Five replicates were used to calculate the average value.

Water contact angles (WCA) of the samples were carried out using an optical contact angle measuring unit (OCA, 154EC). Five values were measured, averaged, and reported.

Surface structures of coated paper with Ag@Fe\(_3\)O\(_4\) and high resolution energy dispersive X-ray spectroscopy (EDS) were observed using scanning electron microscopy (SEM, Hitachi Microscope model S-3000 N). A micrograph analysis was performed at 200 kV accelerative voltage. Surfaces of the samples were sputter-coated with gold before measurement.

Measurement of water vapor transmission rate (WVTR) of the paper coated with Ag@Fe\(_3\)O\(_4\) was performed. A sample with 10 mL of distilled water was weighed and kept in a desiccator at 24 °C and 52% RH. The sample was weighed hourly for a total period of 8 h. The WVTR was determined as a slope of linear portion of the weight gained versus time. The water vapor permeability (WVP) was then calculated based on Eq. (1).

\[
\text{WVP} = \frac{\text{WVTR} \times \text{Thickness}}{\Delta P} \tag{1}
\]

where \(\Delta P\) is the difference in partial pressure of water vapor across the sample (kPa).

Water absorption isotherm of the specimen was also measured. The sample with 20 mm \(\times\) 20 mm was kept in the container filled with MgCl\(_2\), NaCl, Mg(NO\(_3\))\(_2\) and K\(_2\)CO\(_3\) at 25 °C for 45 h. The water absorption isotherm was calculated according to the Guggenheim-Anderson-de Boer (GAB) model as described in Eq. (2).

\[
M = \frac{m_0 \cdot C K_w}{(1 - K a_w)(1 - K a_w + C a_w)} \tag{2}
\]

where \(M\), \(a_w\), \(m_0\) and \(C\) and \(K\) represent equilibrium moisture content, relative humidity/100, single layer (g water/g solid) and GAB constant, respectively.

Ag ion and Fe ion release from 2 cm \(\times\) 2 cm sample into food simulants were studied. The food simulants included deionized water, 10% w/v ethanol and 3% w/v acetic acid. The solution of food simulants was changed periodically for the purpose of determining the amount of silver release from a sample as a function of time. The simulant samples were taken and analyzed by atomic absorption spectroscopy (Perkin Elmer, AAnalyst 100) equipped with an air–acetylene burner. The average value was calculated from the values of three replicates. The kinetics of release was studied according to zero order model, first order model, Higuchi model and Korsmeyer–Peppas model as shown in Eqs. (3–6) to investigate the releasing behavior of silver.

**Zero order model**: \(C_t = k_0t\)  \(\tag{3}\)

where \(C_t\) is the amount of is active ingredient \(k_0\) is zero order rate constant and \(t\) is time (hour).

**First order model**: \(\ln C_t = \ln C_o - k_1t\) \(\tag{4}\)

where \(C_t\) is the amount of is active ingredient, \(k_1\) is the first order rate constant and \(t\) is time (hour).

**Higuchi model**: \(\frac{M_t}{M_o} = k_H t^{1/2}\) \(\tag{5}\)

where \(M_t\) is the amount of active ingredient released at time \(t\), and \(M_o\) represents the amount of active ingredient released at equilibrium state and \(k_H\) is Higuchi’s rate constant.

**Korsmeyer–Peppas model**: \(\frac{M_t}{M_o} = k t^n\) \(\tag{6}\)

where \(M_t\) is the amount of active ingredient released at time \(t\), and \(M_o\) represents the amount of active ingredient released at equilibrium state. \(n\) is the release exponent, and \(k\) is the constant value of the active ingredient–composite system.

The value of \(n\) is determined according to the mechanism:

\(n < 0.5\); quasi-Fickian diffusion,
\(n = 0.5\); normal Fickian diffusion,
\(n = 0.5–1.0\); non-Fickian or preposterous transport, and.
\(n = 1.0\); Case II diffusion.

The initial diffusion coefficient \((D)\) can be approximated from Eq. (7), where \(L\) is the sample thickness, \(M_t\) and \(M_{\infty}\) are the amount of active ingredient released at time \(t\) and equilibrium state, respectively.

\[
\frac{M_t}{M_{\infty}} = 4 \left[ \frac{D t}{\pi L^2} \right]^\frac{1}{2} \tag{7}
\]
The paper coated with Ag@Fe$_3$O$_4$ were tested for antimicrobial activity by agar disk diffusion method. The sample with a diameter of 10 mm were placed in contact with Muller Hinton Agar plates seeded with bacterial inoculums. Approximately $10^8$ CFU/mL of bacterial were cultured overnight in Brain heart infusion broth at a temperature of 37 °C for 24 h. Microorganisms tests were Gram-positive bacteria, S. aureus (ATCC25923) and Gram-negative bacteria, E. coli (ATCC25922). The inhibition area was determined at a temperature of 37 °C.

Total soluble solid (TSS), weight loss and vitamin C amount of the tomatoes were tested. Equal weight and size of tomatoes were placed in three types of containers viz., paper box, coated paper box with polymer matrix and coated paper box with 0.15% Ag@Fe$_3$O$_4$. The box with size of 7 cm width, 7 cm length and 7 cm height was made as a tomato packaging. Four tomatoes were packed in each box. The packed tomatoes were kept at 4 °C. The starting weight of the samples were recorded for weight loss measurement. The TSS and vitamin C amount were measured using Refractometer and Iodine titration, respectively.

**Results and discussion**

Characterization of paper coated with Ag@Fe$_3$O$_4$

XRD was used to evaluate the crystalline of AgNPs and Fe$_3$O$_4$ as depicted in Fig. 1a and b. The crystallographic planes of all samples were found at 15.10° (1 1 0), 16.50° (1 1 0) and 22.50° (2 0 0) (Gong et al. 2017; Xing et al. 2018), as shown in Fig. 1a. For paper coated with polymer matrix (the blend of CMC, PVA, ex-SCG and citric acid), the semi-crystalline structure of the CMC/PVA blend was found at 20 of 19.70° (El-Gamal et al. 2018; Atta et al. 2021a, b). The XRD pattern of paper coated with Fe$_3$O$_4$ and 0.15% w/v Ag@Fe$_3$O$_4$ are shown in Fig. 1b. The paper coated with Fe$_3$O$_4$ showed peaks at 20 of 30.20°, 35.25°, 42.40°, 57.41° and 62.60° which corresponded to (220), (311), (400), (511) and (440) crystallographic planes of face-centered cubic Fe$_3$O$_4$ (Dehghan et al. 2022). For paper coated with Ag@Fe$_3$O$_4$, the characteristic peaks of silver (Ag) were also observed at 38.75°, 44.22° and 64.28° which were assigned to (111), (200) and (220) lattice planes (Yeamsuksawat et al. 2021). The unassigned peak of paper coated with Ag@Fe$_3$O$_4$ at 28.83° in Fig. 1a was related to bio-organic phase that occurred on the surface of AgNPs (Gajendran et al. 2019). Dangi et al. 2020 suggested that the bio-reducing agent capped on AgNPs surface presented a crystalline nature of the bio-organic phase. Furthermore, the peaks of other iron oxides could not be observed. The obtained results confirmed that the Fe$_3$O$_4$ did not oxidize during the green synthesis of AgNPs and the coating process. The elements of Ag@Fe$_3$O$_4$ for coating were analyzed by EDX as depicted in Fig. 1c. The EDX spectra showed the Ag, Fe and C elements which were related to AgNPs, Fe$_3$O$_4$ and polymer matrix, respectively.

ATR-FTIR was used to analyze chemical bond and interaction of samples as depicted in Fig. 2. The characteristic peaks of paper coated with the CMC/
PVA blend were found at 840 cm$^{-1}$ (C–C stretching), 1034 cm$^{-1}$ (C–O–C stretching), 1413 cm$^{-1}$ (COO symmetric), 1588 cm$^{-1}$ (Asymmetric COO), 2920 cm$^{-1}$ (CH$_2$ stretching) and 3280 cm$^{-1}$ (O–H stretching). In the presence of Ag@Fe$_3$O$_4$ coating on the paper, the peaks at 568 and 587 cm$^{-1}$ corresponded to the stretching vibration of tetrahedral group of Fe$^{3+}$–O$^{2−}$. The shift of peak position at 3280 cm$^{-1}$ to 3270 cm$^{-1}$ and 2920 cm$^{-1}$ to 2930 cm$^{-1}$ indicated the strong interaction between Ag@Fe$_3$O$_4$ and the polymer matrix. For the case of incorporating citric acid into the CMC/PVA blend containing Ag@Fe$_3$O$_4$, peaks at 1220 and 1710 cm$^{-1}$ were found and assigned to the stretching linkage that was created by carboxylic groups of citric and hydroxyl groups of the polymer matrix (Ounkaew et al. 2018). A shift in absorption bands at 2940–3320 cm$^{-1}$ corresponding to hydrogen bond interaction was also observed (Ghaseminezhad and Shojasoadati 2016).

Morphology of paper coated with Ag@Fe$_3$O$_4$

The microstructures of paper coated with different contents of Ag@Fe$_3$O$_4$ were observed using SEM and shown in Fig. 3. The paper coated with 0.05–0.15%w/v Ag@Fe$_3$O$_4$ showed homogenous dispersion. An increase in Ag@Fe$_3$O$_4$ content to 0.2%w/v resulted in the formation of aggregated particles. This might be due to the powerful magnetic force from the dipole–dipole interaction between Fe$_3$O$_4$ particles at high content and the polymer matrix (Yeamsuksawat et al. 2021). A similar observation was also found in the carrageenan reinforced with hybrid nanoparticles consisting of silica, polyamidoamine, Fe$_3$O$_4$ and AgNPs (Saedi and Rhim 2020).

Antibacterial activity of paper coated with Ag@Fe$_3$O$_4$

Antibacterial activities of paper coated with Ag@Fe$_3$O$_4$ were evaluated using agar diffusion method. The inhibitory zones for *S. aureus* and *E. coli* are shown in Fig. S1 and summarized in Table 1. The paper coated with Ag@Fe$_3$O$_4$ showed a better antibacterial activity for *S. aureus* than for *E. coli*. The *S. aureus* (Gram-positive bacteria) composed of peptidoglycan layer which allowed the antibacterial agents to enter the bacterial cells easily and quickly (López-De-Dicastillo et al. 2012; Wang et al. 2017). The antibacterial efficiencies of the paper coated with Ag@Fe$_3$O$_4$ increased when content of Ag@Fe$_3$O$_4$ increased up to 0.15%w/v. Generally, small metal oxide particles with good dispersion provided a great surface area and resulting strong antimicrobial interactions (Ranjbar et al. 2020). Interestingly, the synergistic behavior of combination between Ag@Fe$_3$O$_4$ and citric acid was found. The inhibition zones of paper coated with Ag@Fe$_3$O$_4$ at 0.15%w/v were larger than those of paper coated with polymer matrix, only 0.15%w/v Ag@Fe$_3$O$_4$ and only ex-SCG. Based on the literature, several mechanisms for antibacterial activities of AgNPs hybrid with Fe$_3$O$_4$ have been proposed. Dehghan et al. (2022) suggested that the Ag@Fe$_3$O$_4$ might provide higher and quicker diffusion of silver ions for inhibition of bacterial growth. Yu et al. (2018) reported that negative charges of bacteria can be easily wrapped by Ag@Fe$_3$O$_4$ via electrostatic attraction. Moreover, a sustained release of silver ions around the bacterial cells can create the interaction with the thiol groups of vital proteins resulting in the inactivation of enzyme proteins and respiration and causing bacterial death (Dehghan et al. 2022). Raza et al. (2016) proposed that Ag@Fe$_3$O$_4$ had high adhesion capability with cell membrane of bacteria and provided
oxygen radicals that can attack bacterial cell membrane and disrupt enzyme, ribosome and lysosome activities. For citric acid, acidulation and chelating were the main effects for antibacterial activities. The acidic ions can damage enzymes and extracellular membrane whereas chelating agents of citric acid can bind and remove essential metal ions for the growth of bacteria (Ounkaew et al. 2018). Moreover, the presence of ex-SCG in samples might assist the antibacterial activities. The use of ex-SCG combined with other bioactive agents such as oregano essential oil (Trongchuen et al. 2018) and citric acid (Ounkaew et al. 2018) exhibited the synergistic effect on antibacterial activities. The main components of ex-SCG such as chlorogenic acid, caffeic acid and other compounds in ex-SCG can inhibit the bacterial RNA polymerase enzyme and disrupt the permeability of outer and plasma membranes.

Tensile property of paper coated with Ag@Fe3O4

Adequate tensile strength of paper packaging is an important property to maintain its integrity during transportation and storage (Li et al. 2022). Tensile strength of paper coated with Ag@Fe3O4 was determined as a function of Ag@Fe3O4 content and shown in Fig. 4. The uncoated paper had a tensile strength of $21.07 \pm 0.70$ MPa. The tensile strengths increased to $50.42 \pm 1.88$ MPa with $0.05\%\text{w/v}$ Ag@Fe3O4 and to $53.52 \pm 0.71$ MPa with $0.15\%\text{w/v}$ Ag@Fe3O4. The paper coated with $0.15\%\text{w/v}$ Ag@Fe3O4 remarkably enhanced tensile strength about 154.01% compared to uncoated paper. These results help explain that the formation of the strong intermolecular hydrogen bonds inside the paper matrix.

### Table 1 Inhibition zones of coated papers

| Coated paper                        | E. coli (mm) | S. aureus (mm) |
|-------------------------------------|--------------|----------------|
| Polymer matrix                      | 2.84 ± 0.12  | 3.19 ± 0.14    |
| Ag@Fe3O4 0.05%w/v                  | 3.02 ± 0.11  | 3.68 ± 0.21    |
| Ag@Fe3O4 0.1%w/v                   | 3.59 ± 0.10  | 4.24 ± 0.01    |
| Ag@Fe3O4 0.15%w/v                  | 4.51 ± 0.29  | 5.06 ± 0.23    |
| Ag@Fe3O4 0.2%w/v                   | 3.58 ± 0.41  | 4.39 ± 0.15    |
| ex-SCG                              | ND           | ND             |
| Ag@Fe3O4 0.15%w/v (without citric acid and ex-SCG) | ND | 1.16 ± 0.19 |

ND not detected
bond between Ag@Fe₃O₄ and the polymer matrix at the optimal proportion can improve the interfacial bonding and increase the tensile strength (Amini et al. 2016). However, the tensile strength decreased when the paper was coated with 0.2%w/v Ag@Fe₃O₄. This was due to excessive addition of metal particles and inadequate polymer matrix as a binder for coating (Jung et al. 2018).

Water vapor permeability and contact angle of paper coated with Ag@Fe₃O₄

One of main functions of food packaging is to maintain moisture in food products during storage. Therefore, a low WVP is required to minimize the moisture transfer from food to the surrounding atmosphere (Tajik et al. 2013). The WVP of paper coated with various contents of Ag@Fe₃O₄ is shown in Fig. 5. The highest WVP of 5.67 ± 0.32 × 10⁻¹⁰ g/m²×kPa×h was observed in the uncoated paper. A high amount of water can move through the porous structure of the uncoated paper, as observed in SEM image (Fig. 3). The paper coated with 0.15%w/v Ag@Fe₃O₄ remarkably decreased the WVP to 2.92 ± 0.16 × 10⁻¹⁰ g/m²×kPa×h. The decrease in number and size of pores by coating can make paper package less permeable to water vapor (He et al. 2021). Compared with the uncoated paper, the WVP of coated paper with Ag@Fe₃O₄ reduced by 37.39%, 37.68%, 40.45% and 48.50% when Ag@Fe₃O₄ content were 0%w/v (paper coated with polymer matrix), 0.05%w/v, 0.1%w/v and 0.15%w/v, respectively. The uniformly dispersed Ag@Fe₃O₄ could form a tortuous path for water vapor transmission. The obtained results were in good agreement with the work by He et al. (2021) who reported that paper coated by CMC/cellulose nanocrystals immobilized silver nanoparticles (CNC@AgNPs). The presence of CNC@AgNPs in the coated paper decreased WVP up to 43.2%.

The hydrophobicity of paper coated with Ag@Fe₃O₄ were measured using water contact angle (WCA) as illustrated in Fig. 6. The WCA of uncoated paper was 32.1° and increased after coating with 0.05–0.2%w/v Ag@Fe₃O₄. This implied that the surface hydrophobicity of paper can be improved by coating with Ag@Fe₃O₄. This high WCA of coated paper samples can be explained based on roughness of a surface (Ounkaew et al. 2021a, b). As described in Cassie’ s theory, the grooves between the pyramid shaped structures on the surface will be occupied by
air and hinder water to fill in the grooves (Kansal et al. 2020). The images of surface roughness from SEM analysis are shown in Fig. 7. The increase of Ag@Fe₃O₄ content in coating resulted in the enhancement of surface roughness. The improvement of hydrophobicity of coated paper would be beneficial for a wide range of applications.

Moisture sorption isotherms of paper coated with Ag@Fe₃O₄

During storage or in service, the cellulose-based materials used for packaging have moisture absorption or desorption from and to the surrounding air (Wang et al. 2018). The presence of water in packaging influences its properties such as mechanical and barrier properties (Mohammadi Nafchi et al. 2014). The moisture sorption isotherms of paper coated with polymer matrix and 0.15%w/v Ag@Fe₃O₄ are illustrated in Fig. 8. Two different regions of equilibrium moisture content (EMC) for all samples were observed. Slow moisture sorption was found at water activity of 0.32–0.52, and then rapid moisture sorption was observed at high water activity of 0.52–0.75. This behavior indicated the surface adsorption of water vapor by cellulose-based materials such as paper-based food packaging (Irimiaia 2021), paperboard (Rhim 2010) and paper sheets (Mihaly-Cozmuta et al. 2017). The paper coated with 0.15%w/v Ag@Fe₃O₄ can minimize moisture sorption with lower EMC value compared to the paper coated with polymer matrix. This observation was in good agreement with the results of WCA and WVP results. Furthermore, the moisture sorption isotherm parameters of uncoated and coated paper were also determined using GAB model. The obtained parameters from GAB model are summarized in Table 2. The incorporation of 0.15%w/v Ag@Fe₃O₄ in coating resulted in a reduction of a monolayer value indicating the maximum absorbed water content in a single layer per gram of dry material. Bedane et al. (2015) suggested that water molecules in the multilayers behaved like a pure liquid when the K value is close to 1. The increase in K value for paper coated with Ag@Fe₃O₄ implied the less structured configuration of water molecules adsorbed in the multilayer. The obtained C values of paper coated with polymer matrix and 0.15%w/v Ag@Fe₃O₄ were consistent with the values found in previous studies involving cellulosic material and biopolymer; cellulose film from bleached kraft pulp (Bedane et al. 2015), and carboxymethyl cellulose (Torres et al. 2012).
Release of metal ions from Ag@Fe₃O₄ in coated paper to food simulants

Release of Ag ion can effectively inhibit the growth of bacteria (Xiong et al. 2013; Lu et al. 2017). Yu et al. (2018) suggested that the Ag ion release from Ag@Fe₃O₄ hybrid was an important parameter for antibacterial activities. Moreover, a toxicity of migrating Ag ions from AgNPs in food packaging is still a great concern (Srikhao et al. 2021). According to the EU Regulation 10/2011 (European Union 2011), the overall migration limit (OML) of permitted compounds from plastic to contact with foodstuff was suggested at 60 mg of substances/kg of food simulants or food packaged (Agustinelli et al. 2021). Different kinds of food simulants were used to test the release of silver ions. Three food simulants, viz, deionized water, 3% w/w acetic and 10% w/w ethanol were prepared to represent the polar, low pH and alcoholic...
The migration of Ag ions from the paper coated with Ag@Fe₃O₄ in three food simulants is shown in Fig. 9(A). After immersion of samples in food simulants for 168 h, the cumulative Ag ion release ranged from 0.076 to 1.30 mg/kg of food simulants which was lower than the OML value. The release capability of silver in the three food simulants was in descending order as follows; 3% w/w acetic acid > 10% w/w ethanol > distilled water. The AgNPs had a higher solubility in acid than the other organics. The damaged bonds of Ag₂O on AgNPs surface in an acidic environment also enhanced the Ag ion release (Lu et al. 2017; Agustinelli et al. 2021). In addition, the hydrophilic nature of Ag@Fe₃O₄ allowed water molecules to penetrate into the sample. The hydrated sample resulted in the segregated polymer matrix which caused more migration of Ag ions (Srikhao et al. 2021).

Fe₃O₄ is approved by the Food and Drug Administration for drug delivery and nutritional supplement usage. According to the Nordic Nutrition Recommendations, iron intake is recommended depending on gender and age. For example, the recommended iron intakes for boys (age 14–17 years), men (age 18 years and older) and women of the childbearing age are 11 mg iron/day, 9 mg iron/day and 15 mg/day, respectively. The releases of Fe ions from the paper coated with Ag@Fe₃O₄ in the three food simulants are shown in Fig. 9(B). The cumulative Fe ion release was 3.09 mg/kg of food simulant for 3% w/w acetic acid, 2.10 mg/kg of food simulant for distilled water and 2.46 mg/kg of food simulant for 10% w/w ethanol. The Fe ion release of paper coated with Ag@Fe₃O₄ did not exceed the recommended iron intake. The highest release was found with 3% w/w acetic acid. In acidic solution, the water-insoluble Fe₃O₄ was reduced to be ferrous ion which was soluble in water.

The release kinetic of metal ions from the paper coated with Ag@Fe₃O₄ in three food simulants was also investigated. Four different release mathematical models viz., zero order, first order, Higuchi and Korsmeyer-Peppas models were applied to determine kinetic release parameters. The release profiles of Ag and Fe ions are shown in Fig. S2. The Korsmeyer-Peppas model showed a better fit for the release of Ag ions in three food simulants with higher R² values. For the release of Fe ions, the release profile in 3% w/w acetic acid was the only one that could be fitted with Korsmeyer-Peppas model. The releases of Fe ions in distilled water and 10% w/w ethanol were quite constant over the release period. As shown in Fig. 9b, the less soluble Fe ions in distilled water and 10% w/w ethanol were found at 2 h, and then no further release was observed. The kinetic release parameters are summarized in Table 3. The values of n for the release of Ag and Fe ions were lower than 0.5 which indicated a quasi-Fickian diffusion mechanism. The release of metal ions was controlled by the corrosion of metal at the grain boundaries by food simulant.

![Fig. 9](image-url) Release of a Ag ions and b Fe ions from coated paper with 0.15% w/v Ag@Fe₃O₄ into food simulants.
(Cano et al. 2016). Furthermore, the diffusion coefficient values of Ag and Fe ions showed variation owing to the different natures of food simulants.

Application of paper coated with Ag@Fe₃O₄ for tomato packaging

The uncoated paper and paper coated with polymer matrix and 0.15%w/v Ag@Fe₃O₄ were used to package tomatoes. The changes in weight loss, TSS content, and vitamin C content of tomatoes are shown in Fig. 10a–c. As shown in Fig. 10a, the weight loss of tomatoes in the uncoated paper was higher than those of paper coated with polymer matrix and 0.15%w/v Ag@Fe₃O₄ over storage time. The lowest weight loss was found with the paper coated with 0.15%w/v Ag@Fe₃O₄. The increase in hydrophobicity of coated paper decreased moisture and gas permeability, respiration rate and water loss (Ounkaew et al. 2021b; Pagliarulo et al. 2016).

TSS is a parameter indicating the sweet flavor in fruits and vegetables (Ounkaew et al. 2021a; Jiang et al. 2019). Figure 10b exhibits the TSS values of samples. The TSS of all samples increased throughout storage. The increase in TSS can be explained by several factors such as a breakdown of starch to sugar (Ashurst and Arthey 2000), a reduction of respiration rate (Ghaseminezhad and Shojaosadati 2016), and loss of water (Dris et al. 1999). The coated paper can lower the TSS value and the lowest value was found in the paper coated with 0.15%w/v Ag@Fe₃O₄. A lower TSS implied a slower rate of hydrolysis of carbohydrates, thus prolonging the storage life of fruit and vegetable (Tajik et al. 2013).

Tomato is considered as a major dietary source of vitamin C and lycopene (Badin et al. 2021). Figure 10c shows the vitamin C content of tomatoes in three different packaging. For uncoated paper and paper coated with polymer matrix, vitamin C increased during the first 5 days of storage and then

| Metal ions release | Food simulants | Korsmeyer-Prppas |  |  |
|-------------------|---------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|                   |               | Correlation coefficient (R²) | Release Exponent (n) | Release Factor (k) | Diffusion Coefficient (D×10⁻³), mm²/h |
| Ag ions           | Deionized water | 0.9841 ± 0.014 | 0.218 ± 0.182 | 0.459 ± 0.432 | 1.67 ± 0.151 |
|                   | 10%wt ethanol  | 0.9856 ± 0.007 | 0.281 ± 0.190 | 0.346 ± 0.323 | 2.47 ± 0.22 |
|                   | 3%wt acetic acid | 0.9980 ± 0.017 | 0.264 ± 0.142 | 0.324 ± 0.221 | 4.7 ± 0.32 |
| Fe ions           | Deionized Water | 0 | – | – | – |
|                   | 10%wt ethanol  | 0 | – | – | – |
|                   | 3%wt acetic acid | 0.9953 ± 0.004 | 0.093 ± 0.016 | 0.622 ± 0.050 | 0.2 ± 0.01 |

Fig. 10 Quality of tomatoes in uncoated paper and paper coated with polymer matrix and Ag@Fe₃O₄ packaging a Weight loss, b TSS value and c Vitamin C content
declined towards the end of storage. The use of paper coated with 0.15%w/v Ag@Fe₃O₄ showed an increase in vitamin C over the storage time because the tomato gradually matured during storage. The decrease in vitamin C might be attributed to external oxidation (He et al. 2021) and low respiration rate (Brasil et al. 2012). Zhang et al. (2020) suggested that AgNPs in polylactic acid films could change membrane permeability, inhibit respiration, and delay the oxidation reaction of ascorbic acid in fruits. Hence, applying the paper coated with 0.15%w/v Ag@Fe₃O₄ for packaging is beneficial in maintaining essential nutrients of food products.

Figure 11 exhibits the appearance change of tomatoes packed in paper packaging. The tomatoes in the uncoated paper showed wrinkled skins whereas the slight wrinkle skins were observed for tomatoes packed in paper coated with polymer matrix and 0.15%w/v Ag@Fe₃O₄. To achieve the extended shelf-life and quality of tomatoes, the paper coated with 0.15%w/v Ag@Fe₃O₄ would be a good candidate for use as bioactive packaging.

**Conclusion**

In this research, Ag@Fe₃O₄ was synthesized using eco-friendly technique for coating packaging paper for food preservation. The CMC/PVA crosslinked by citric acid was used as polymer matrix. The result from the antibacterial test showed that the paper coated with 0.15% Ag@Fe₃O₄ exhibited the highest inhibition zone for both *S. aureus* and *E. coli*. Moreover, some synergistic effects on mechanical properties of polymer matrix and Ag@Fe₃O₄ were observed. The tensile strength of paper coated with Ag@Fe₃O₄ increased with increasing Ag@Fe₃O₄ in the range of 0.05–0.15%w/v due to the good dispersion of 0.15%w/v of Ag@Fe₃O₄ in the polymer matrix. The WVP of paper coated with 0.15%w/v Ag@Fe₃O₄ was reduced by 48.50% compared to that of the uncoated paper. Furthermore, WCA of the paper coated with Ag@Fe₃O₄ showed greater hydrophobicity of the samples. The release of Ag ions was lower than the minimum specified by EU Regulation 10/2011 whereas the release of Fe ions was lower than the maximum allowable human intake. Interestingly, the paper coated with 0.15%w/v Ag@Fe₃O₄ can be applied for bioactive packaging as it can effectively extend shelf-life, reduce weight loss and retain vitamin C of tomato.
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