Characterization and properties of LDPE film with gallic-acid-based oxygen scavenging system useful as a functional packaging material

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ABSTRACT: We prepared and characterized active, oxygen-scavenging, low density polyethylene (LDPE) films from a non-metallic-based oxygen scavenging system (OSS) containing 1, 3, 5, 10, and 20% of gallic acid (GA) and potassium chloride (PC). We compared the surface morphology and mechanical, permeability, and optical properties of the oxygen-scavenging LDPE film with those of pure LDPE film. The surface morphology, gas barrier, and thermal properties indicate that the OSS was well incorporated into the LDPE film structure. The surface roughness of the film increased with the amount of oxygen scavenging material. The oxygen and water vapor permeability of the developed film also increased with the amount of oxygen scavenging material, though its elongation decreased. The oxygen scavenging capability of the prepared film was analyzed at different temperatures. The initial oxygen content (%) in the vial headspace, 20.90%, decreased to 16.6% at 4 °C, 14.6% at 23 °C, and 12.7% at 50 °C after 7 days of storage with the film containing 20% OSS. The film impregnated with 20% organic oxygen scavenging material showed an effective oxygen scavenging capacity of 0.709 mL/cm^2 at 23 °C. Relative humidity triggered the oxygen scavenging reaction. © 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 44138.

INTRODUCTION

Residual oxygen in the headspace of a package can cause the oxidation of packaged food and the growth of aerobic bacteria or molds.[1–4] Vacuum packaging, modified atmosphere packaging, and oxygen scavenging technology are some of the most commonly used methods to remove or reduce headspace oxygen in the food packaging industry.[5] Among those three techniques, only oxygen scavenging can reduce and maintain headspace oxygen content of less than 1% during storage.[6] A variety of oxygen scavengers have been commercialized for use in the food packaging industry. These oxygen scavenging systems are used in various forms, such as sachets, plastic films, labels, plastic trays, and bottle crowns.[7]

Iron powder has been extensively used as a commercial oxygen scavenging material because of its cost and oxygen absorption capability. However, iron-based oxygen scavenging sachets have several disadvantages. They pose a health risk to consumers by accidental ingestion, and they cannot be used for liquid products.[3,5,8,9] In addition, a package containing iron-based oxygen absorbers should not be heated in a microwave oven because

ignition can result, and iron-based oxygen absorbers are detected by the metal detectors often used in packaging lines.[10] For those reasons, active packaging research currently focuses on natural compounds as the basis for oxygen scavengers. Several researchers added an oxygen scavenger into a polymer matrix.[11–13] These active oxygen scavenging films effectively scavenged oxygen and could be very useful in food packaging.

Using natural compounds for oxygen scavenging could provide several benefits.[7] The consumers perceptions of a natural food is very positive and there for use of natural based oxygen scavenger in package will add benefit.[14] The compounds have already been incorporated into polymer materials as a stabilizer and antioxidant to reduce oxidation in food products.[15,16] In addition, they do not set off metal detectors, and they carry no possibility of ignition, even if heated in a microwave oven.[10]

Gallic acid (2,3,4-trihydroxybenzoic acid) is a naturally occurring plant phenol that acts a strong free radical scavenger and is commonly used to prevent lipid oxidation in processed food. The use of natural phenolic compounds in food packaging is particularly encouraged because they improve food oxidative

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Table I. Composition of Each Oxygen Scavenging Film

Sample	Materials			Percentage of scavenging materials added to the polymer
	LDPE resin (g)	Gallic acid (g)	Potassium carbonate (g)	
LDPE	300	—	—	0
OSS 1%	297	2	1	1
OSS 3%	291	6	3	3
OSS 5%	285	10	5	5
OSS 10%	270	20	10	10
OSS 20%	240	40	20	20

Abbreviation: OSS, oxygen scavenging system.

and microbial status and show many different benefits for human health.[17] Gallic acid oxidizes with the formation of hydrogen peroxide, quinones, and semi-quinones in an alkaline environment.[18] Gallic acid is, therefore, an effective natural compound for oxygen scavenging.

Potassium carbonate (K_2CO_3) has been used to absorb carbon dioxide.[19] It forms a strongly alkaline solution when dissolved in water and can be used as the catalyst for gallic-acid-based oxygen scavenging.

The objectives in this research were to evaluate the oxygen scavenging capability of gallic acid and potassium carbonate based LDPE film. We incorporated the gallic-acid-based oxygen scavenger into low density polyethylene (LDPE) film and tested the influence of moisture on the oxygen scavenging capability of the system at 4 °C, 23 °C, and 50 °C. We also studied the surface morphology and mechanical, barrier, and optical properties of the film.

EXPERIMENTAL

Materials
Gallic acid monohydrate (Fw188.14) was purchased from Kanto Chemical Ltd. (Portland, OR, USA). Potassium carbonate anhydrous (99.5%), lithium chloride anhydrous (98.0%), and magnesium chloride anhydrous (98.0%) were purchased from Dae-Jung Co. (Kyungki, Korea). Magnesium nitrate hexahydrate (99.0%) and ammonium sulfate (99.5%) were purchased from Junsei Ltd. (Tokyo, Japan), and LDPE (Lutene LB7500 LDPE) was purchased from LG Chemical Information & Electronic Materials Co. (Nanjing, China).

Preparation of Oxygen Scavenging Films
Conditioning of Materials. The test samples to remove the moisture from the materials were pre-conditioned by drying them under vacuum at 105 °C for 12 h before performing a pre-film extrusion process.

Preparation of Film. The oxygen scavenging film containing 1, 3, 5, 10, and 20% of the oxygen scavenging system (OSS) (Table I) were manufactured using a laboratory extrusion cast film line (Compounding Extruder, Bautek Co., Gyeonggi, Korea) equipped with two screw extruders (L40/D19), a flow convergence system (feed-block), a coat-hanger type head (slit die of 200 3 0.25 mm²),

and a take-up/cooling system (chill rolls) thermally controlled by water circulation at 10 °C. The temperature of the extruder was divided into seven zones. We prepared the film in the 110 °C Zone 7 (feed zone). The header was set at 160 °C in Zones 1–6 (Metering and Compression zones). The pressure of the twin screw extruder barrel was 8.4 kgf/cm².

Determination of Film Properties
Mechanical Properties. The tensile strength (TS) and elongation at break (%E) of the films were measured using an Instron Universal Testing Machine (Daekyung Tech & Tester CO, Seoul, Korea) according to ASTM standard D882-91.[20] Specimen samples 10 cm × 2.54 cm were cut from developed film and prepared on glass plates. Specimen samples conditioned for 48 h at 23 °C and 50% relative humidity (RH) in a constant temperature and humidity chamber before the measurement of mechanical properties of film. The grip separation was 50 mm and extension rate was maintained at 500 mm/min. The values are the average of six measurements.

Color Measurements. Film color was measured using a color difference meter (CR-400, Minolta Camera Co., Osaka, Japan). The instrument with a standard white plate was calibrated and used the (CIE) Lab color space (CIE, 1986). Lightness (L^*) and chromaticity parameters a^* (red–green) and b^* (yellow–blue) were obtained using illuminant D65 at an observer angle of 2° (Moreno-Osorio et al., 2010). Five measurements were taken from each sample using three samples of each film. Total color difference (ΔE) was calculated according to eq. (1).

$$\Delta E = \sqrt{(L_0^* + L^*)^2 + (a_0^* + a^*)^2 + (b_0^* - b^*)^2} \qquad (1)$$

Where L_0^*, a_0^*, and b_0^* are the value of pure LDPE film, and L^*, a^*, and b^* are the measured values of the samples.

Oxygen Transmission Rate of Films (OTR). The OTR of the films was measured according to ASTM standard D3985[21] by using an 8001 oxygen permeation analyzer (Illinois Instruments Co., Johnsburg, IL, USA). Oxygen transmission rates were set with an auto stop program when the rate of error was ±1%. The flow rates of oxygen and nitrogen were 20 and 10 cm³/min, respectively. Samples were exposed to 50% RH and tested at 23 ± 1 °C. Oxygen permeability (g mm/m² h kPa) was calculated by multiplying the oxygen transmission rate by the film thickness. The test was in triplicate and reported the mean value. The OTR of the film was calculated according to eq. (2).

$$\mathrm{OP}\left(g \cdot mm / m^2 \cdot h \cdot kPa\right) = \frac{OTR \cdot T}{\Delta P} \qquad (2)$$

Where OP is oxygen permeability coefficient, g mm/m² h kPa; OTR is the oxygen transmission rate, g/m²·day; T is thickness, mm; $\triangle P$ is the partial pressure difference across the film wall, kPa.

Water Vapor Permeability (WVP). The WVP coefficient of the films was measured according to ASTM standard F-1249 [22] by using a 7001 water permeation analyzer (Illinois Instruments Co. Johnsburg, IL, USA) by cutting a 500 × 500 mm² film sample. The samples were exposed to 50% RH and tested at (23 ±1) °C. The data were directly obtained from the phosphorous pentoxide (P_2O_5) moisture sensor. Water vapor permeability (WVP; g mm/m² h kPa) was calculated by multiplying the water transmission

rate by the film thickness. The test was in triplicate and reported the mean value. The water vapor permeability coefficient of the film was calculated according to eq. (3).

$$\text{WVP}\left(g \cdot mm/m^2 \cdot h \cdot kPa\right) = \frac{WVTR \cdot T}{\Delta P} \qquad (3)$$

Where WVTR is the water vapor transmission rate, $g/m^2 \cdot day$; T is thickness, mm; and $\triangle P$ is the partial pressure difference across the film wall, kPa.

Characterization of the Film

Morphological Structure. The surface and cross section morphologies of the film samples were examined using a field emission scanning electron microscope (SEM). The film samples were fractured in liquid nitrogen and coated with a fine gold layer before obtaining the micrographs. All samples were examined at a voltage of 5 kV. (Field Emission Scanning Electron Microscopy, JSM-6700F, JEOL Co., Freising, Germany).

Fourier Transform Infrared (FTIR) Spectroscopy. FTIR spectroscopy was performed to observe the structural interactions between the LDPE films and the gallic acid-potassium carbonate OSS. The FTIR spectra of the films were recorded from 4000–450 cm^{-1} at a resolution of 16 cm^{-1} using an FTIR spectrometer (PerkinElmer, Billerica, MA, USA).

Thermal Properties. The thermal properties of the oxygen scavenging film were analyzed using a differential scanning calorimeter (DSC; DSC-Q10, TA Instrument Co., Wood Dale, IL, USA) and thermo-gravimetric analyzer (TGA; TGA-4000, PerkinElmer Co., Groningen Netherlands). For DSC, the films were scanned from $-30\,^\circ$C to $-400\,^\circ$C at a rate of 10 $^\circ$C/min. For TGA, films were scanned from 20–700 $^\circ$C at a rate of 20 $^\circ$C/min. Nitrogen was used as the purge gas at a flow rate of 20 mL/min for both DSC and TGA.

Oxygen Scavenging Capability of Film

The film samples were cut into two pieces (8 $\times$ 6 cm^2, weight 4.0 g). The films were folded and placed them in a clear 115 mL headspace vial as shown in Figure 1, with 115 mL of ambient air (20.90%, O_2). The final glass vials were prepared with 0.5 mL of water to generate 95% RH and sealed with 20 mm polytetrafluoroethylene/silicone septa. The sealed vials were stored at different storage temperatures. For this study 4, 23, and 50 $^\circ$C temperature were selected, because commonly, we store food in three different storage conditions that is low, medium, and high temperature. Hence we selected three different temperature 5 $^\circ$C (cold storage temperature), 23 $^\circ$C (room temperature), and 50 $^\circ$C (from this temperature lipid oxidation starts in food products) and determined how LDPE/GA film affect the oxygen scavenging capability in different storage temperature. The storage temperature and RH inside the glass vials were analyzed using a thermos-hygrostat (Model SK-L200THIIα, Sato, Co., Tokyo, Japan).

The oxygen scavenging capability of the prepared films was determined according to the method described by Shin et al.[23] The amount of oxygen in each vial was measured using an oxygen/carbon dioxide analyzer (PBI-Dansensor America Inc., Glen Rock, NJ, USA). A sampling needle with a 0.45 mm PTFE filter was inserted and sampled 15 mL of headspace gases through a
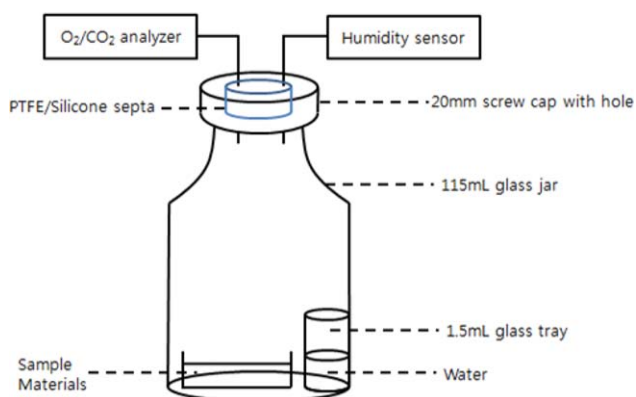

Figure 1. Schematic representation of oxygen scavenging capability testing apparatus. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

septum. The headspace analyzer was calibrated using ambient air after each sample measurement. The test was performed in triplicate and report the mean value.

Statistical Analysis

Data for the experimental results was performed with an analysis of variance (ANOVA) using the SPSS software (SPSS 10.0 for Windows, SPSS Inc. Chicago, IL, USA). The statistical significance of differences between mean values was established as $P < 0.05$ and applied Duncan's New Multiple Range Test for all statistical analyses.

RESULTS AND DISCUSSION

Properties of Film

Mechanical Properties. The TS and %E for the different concentrations of non-metallic oxygen scavenger incorporated into pure LDPE films are shown in Figure 2. The incorporation of the non-metallic OSS caused a reduction in both measurements compared with the control LDPE film. The TS and %E of developed LDPE with OSS decreased with increase in OSS containing gallic acid and potassium carbonate content. Sun et al.[24] reported a decrease in %E values in chitosan-gallic acid films, which indicated that the incorporation of gallic acid into the chitosan film resulted in a strong reaction between the filler and matrix that decreased %E by restricting the motion of the
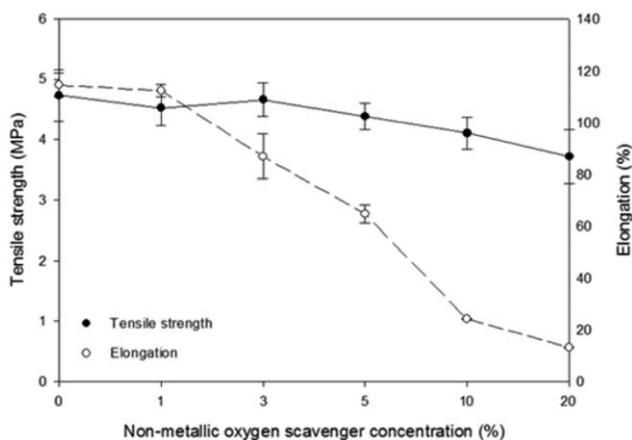

Figure 2. Mechanical properties of the respective films impregnated with different amounts of oxygen scavenging materials.

Table II. Effects of Oxygen Scavenging Material Content on the L, a, b, and $\triangle E$ Values of the Films

Sample	L	a	b	$\triangle E$
LDPE	96.46 ± 0.06^a	-0.59 ± 0.06^a	1.79 ± 0.06^a	0.37 ± 0.04^a
OSS 1%	96.56 ± 0.06^a	-0.62 ± 0.06^a	1.79 ± 0.06^a	0.40 ± 0.06^a
OSS 3%	93.26 ± 0.42^b	-0.16 ± 0.00^a	4.49 ± 0.15^b	4.13 ± 0.41^b
OSS 5%	88.76 ± 0.85^c	1.61 ± 0.17^b	3.69 ± 0.20^{bc}	8.15 ± 0.92^c
OSS 10%	76.16 ± 0.93^d	7.01 ± 0.03^d	8.95 ± 0.38^d	22.71 ± 0.96^d
OSS 20%	71.16 ± 0.95^e	4.54 ± 0.49^c	4.05 ± 0.78^c	25.88 ± 1.16^e

Abbreviation: OSS, oxygen scavenging system.
Values are given as mean $\pm$ standard deviation. Different letters in the same column indicate significant differences ($P < 0.05$) when analyzed by Duncan's New Multiple Range Test.

matrix. The addition of a non-metallic OSS based on α-tocopherol decreased TS and increased %E in gelatin film.[18] The decrease in the tensile strength in the developed LDPE added with OSS films can be because of the uneven dispersion of gallic acid and potassium carbonate in the LDPE matrix. In synthetic polymer blends, the addition of the immiscible component to a ductile matrix generally decreases the elongation properties at break. The elongation would therefore depend on the state of the interface [25] Similar to tensile strength, decrease of percentage of elongation occurred because of the weak interfacial adhesion between LDPE and fillers. Thus, mechanical performance of a filled polymer depends on the strength and the filler module, which further explains the lower tensile strength in our developed film. Particle agglomeration tends to reduce the strength of a material because the agglomerates are weak point that break easily under stress.[23] Elongation of film is decreased, hence we can conclude that structural integrity compromised the film's structural integrity.

Color Measurements. The color, expressed as L, a, b, and ΔE values, of the oxygen-scavenging LDPE film with 1, 3, 5, 10, and 20% of OSS is shown in Table II. As the level of OSS increased, we observed higher a, b and ΔE values with concomitant lower values of L^* ($P < 0.05$). Values of L, a, b, and ΔE for the oxygen scavenging film containing 20% OSS were 71.16 ± 0.95, 4.54 ± 0.49, 4.05 ± 0.78, and 25.88 ± 1.16, respectively. The L value of the oxygen scavenging film decreased from 96.46 ± 0.06 to 71.16 ± 0.95, but the value of a increased from -0.59 ± 0.06 to 4.54 ± 0.49, and b increased from 1.79 ± 0.06 to 4.05 ± 0.78, indicating a tendency toward redness and yellowness compared with the control LDPE film. Thus, the incorporation of the OSS into the LDPE film significantly affected the L and a parameters. The color changes in the LDPE films were most likely caused by the phenolic content of the gallic acid. Phenolic compound added in the films, attributable to chemical moieties and amount of pigments in phenolic compounds. Our results agree with those of Phakawat et al.,[26] who reported that fish skin gelatin film that incorporated root essential oils with phenolic compounds showed high yellowness.

Oxygen Transmission Rate and Water Vapor Permeability. The oxygen permeability coefficient for the different concentrations of non-metallic OSS is shown in Figure 3. Adding gallic-acid-based oxygen scavenger to the LDPE film structure increased the oxygen permeability over that of pure LDPE film. Pure LDPE film with

the addition of 1, 3, 5, 10, and 20% of OSS showed 761.08, 796.91, 989.56, 1092.74, and 1229.27 g mm/m^2 kPa oxygen permeability coefficient, respectively. The higher OTR of the oxygen scavenger film could be caused by the rearrangement of molecules that follows the addition of oxygen scavenging chemicals into LDPE. The developed film does not qualified for good oxygen prevention properties compared with the pure LDPE film. The high OTR value of film containing 10% and 20% OSS might be because of the non-cross-linking gallic acid particles scattered in the film which may have decreased the intermolecular forces between polymer chains, thus increasing the free volume and segmental motions and resulting in the formation of pores. The free volume size plays a crucial role in barrier properties with a linear correlation between oxygen permeability and free volume size.[27] Poor miscibility increases the free volume in a film structure. The addition of gallic acid and potassium carbonate to LDPE film induced a larger free volume in the film structure. Oxygen permeability thus increased because the LDPE film matrix was interrupted.

The WVTR of all films produced in this study is also shown in Figure 3. A significant difference was observed in the measured WVP value between the films containing oxygen scavenger and the control LDPE film. The moisture permeability of the 0, 1, 3, 5, 10, and 20% OSS-impregnated films was 0.87, 0.93, 0.96, 1.22, 1.28, and 1.3827 g.mm/m^2·h·kPa, respectively. This effect was caused by the carboxyl groups and hydroxyl groups of gallic acid are hydrophilic groups, which might promote water transfer in the matrix.[28] DSC analysis also confirm that presence of hydroxyl
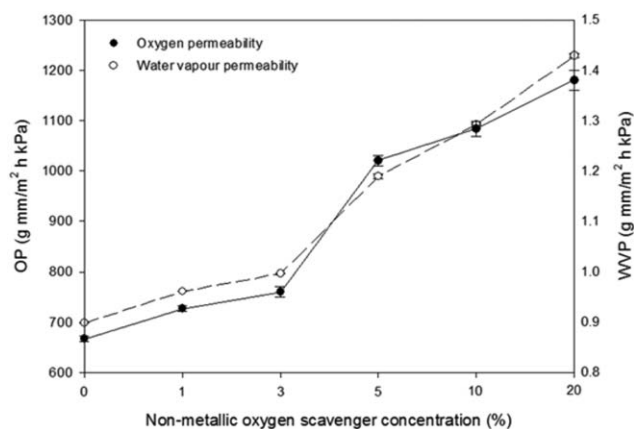

Figure 3. Water vapor and gas permeability's of the respective films impregnated with different amounts of oxygen scavenging materials.
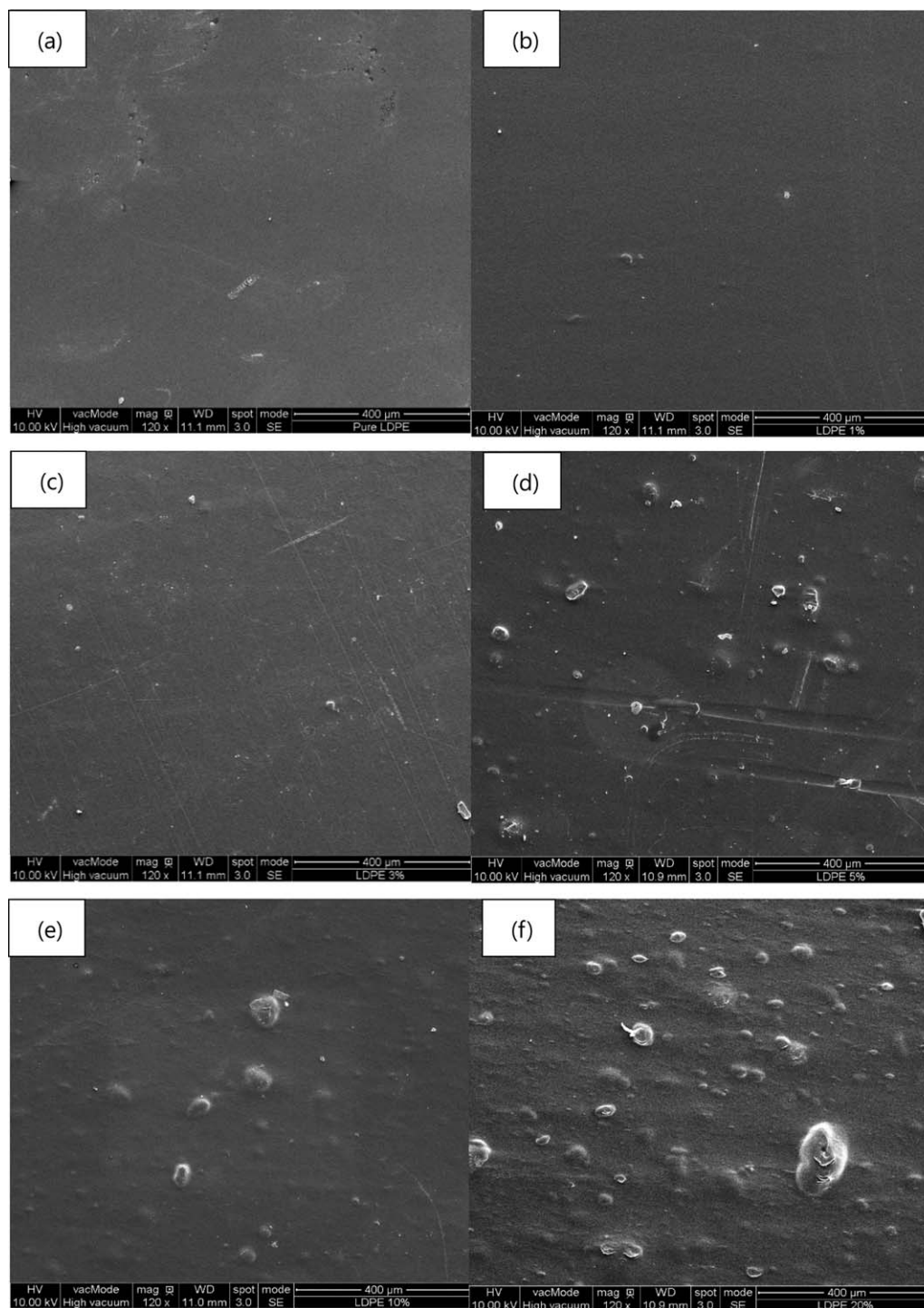
Figure 4. SEM photographs of (a) pure LDPE film, scale bar = 400 μm; (b) LDPE film with 1% OSS, scale bar = 400 μm; (c) LDPE film with 3% OSS, scale bar = 400 μm; (d) LDPE film with 5% OSS, scale bar = 400 μm; (e) LDPE film with 10% OSS, scale bar = 400 μm; (f) LDPE film with 20% OSS, scale bar = 400 μm.

group in the developed LDPE film containing OSS. These results are in agreement with those obtained by Matche et al.,[29] who reported that the difference in WVTR between oxygen scavenging film and pure LDPE could result from the rearrangement of molecules caused by the combination process. Gallic-acid-based oxygen scavenging films are sensitive to moisture. The hydrophilic properties of gallic acid have important role in water barrier properties of the film.[30] The addition of an oxygen scavenging system composed of α-tocopherol nanoparticles enhanced the WVP of fish gelatin films.[31] The authors assumed that a certain degree of film matrix disruption during storage time caused the decrease in the barrier properties of the composite film. In our
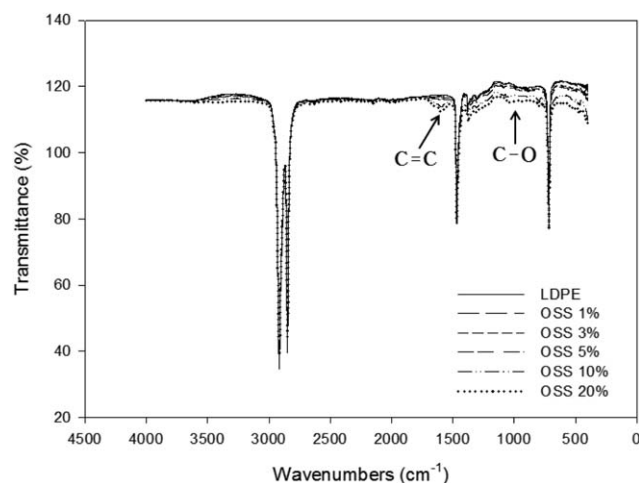
Figure 5. FTIR spectra of the films impregnated with different amounts of oxygen scavenging materials.

Figure 6. TGA curves of the films impregnated with different amounts of oxygen scavenging materials.

research, the gallic-acid-based oxygen scavenging film had higher water vapor properties than the pure LDPE film.

Characterization of the Film

Morphology. Figure 4 shows SEM images of the LDPE films with OSS. The surface of the control LDPE film appears homogeneous with no bubbles or particles, whereas particles appear on the surfaces of the films containing OSS. In addition, the number of particles on the surface increased with the amount of OSS added. The incorporation of phenolic compounds increased film porosity.[32] A micrograph of active gelatin-based films displayed a heterogeneous surface and porous formation after the addition of antioxidant extracts.[33] However, the addition of the gallic-acid-based OSS did not result in film porosity. This result suggests that gallic acid did not release during film formation. We predicted that the OSS in LDPE films would absorb oxygen in a package. The morphological difference in the films could account for the change of WVP obtained in the films. It is possible that the excessive gallic acid cracked the inner structure of the film.[24]

FTIR Spectroscopy. FTIR spectroscopy to inspect the interaction between the LDPE film and the gallic acid-potassium carbonate was used by measuring the absorbance in the wavenumber range of 4000–450 cm^{-1} at a resolution of 16 cm^{-1} (Fig. 5).

The addition of the OSS did not have much effect on the peak; almost all the film peaks have a similar pattern. The control LDPE and OSS-containing film showed the highest peaks at 2900–2800 cm^{-1} for (CH) stretching. The peaks at 722 cm^{-1} and 1911 represent the skeletal vibrations of CH$_2$. Lee et al.[34] reported that gallic acid has a carboxylic group that is activated and converted from an acid to an ester group. Gallic acid could be conjugated at C-3 and C-6 to obtain an ester linkage.[24] The spectra of the control, 1, 3, 5, 10, and 20% OSS LDPE films showed significant peaks around 700 cm^{-1} and at 1400 cm^{-1} caused by the ester group in the gallic acid. A high peak at 2900 cm^{-1} found in all the samples, corresponds to the CH$_2$ asymmetric stretch. Two peaks at 1000 cm^{-1} and 1591 cm^{-1}, called the C–O and C═C peaks, respectively in 20% OSS containing film which confirms the high gallic-acid content of that sample. Potassium carbonate with carbonyl groups display peaks around 1750. These results
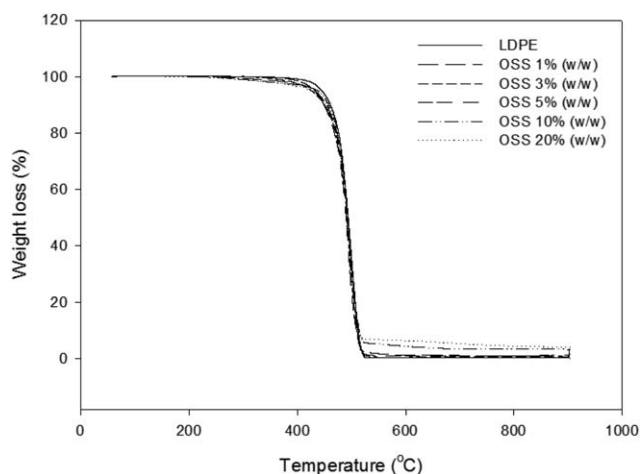
show that the gallate group of the gallic acid was successfully cross-linked with the LDPE via ester linkages for the OSS 1, 3, and 5% samples, but the OSS 20% contained extra gallic acid.

Thermal Properties. The thermal properties of the control LDPE and LDPE with OSS were analyzed using TGA (Fig. 6). The temperature characteristics of weight loss and the melting points (T_m) measured by DSC are summarized in Table III. The T_m of the LDPE film as measured by DSC is 106.3 °C, and 10, 30, and 50% weight loss were observed at 461, 483, and 494 °C, respectively. More than 68% degradation of gallic acid takes place in a temperature range of 68–375 °C, as reported by Santos et al.[35] Gallic acid undergoes a facile thermal decarboxylation to produce pyrogallol. Thus, the second event, at 213 °C, corresponds to the liberation of carbon dioxide upon heating. The third event, at 316 °C, probably corresponds to the further loss of hydroxyl groups. Finally, the fourth peak corresponds to the residual decomposition of gallic acid. The phase change of potassium carbonate takes place in a temperature range of 880–1000 °C. Thermal decomposition does not occur below 1000 °C.[36] The results show that adding gallic acid and potassium carbonate to the LDPE film reduced the film's thermal stability.

Table III. Thermal Properties of the Films Impregnated with Different Amounts of Oxygen Scavenging Materials

Sample	Thermal properties (°C)			
	T_m[a]	$T_{10\%}$[b]	$T_{30\%}$[c]	$T_{50\%}$[d]
LDPE	106.3	461	483	494
OSS 1%	106.5	459	482	492
OSS 3%	106.6	458	482	492
OSS 5%	106.6	455	482	492
OSS 10%	106.7	453	481	491
OSS 20%	106.7	453	480	491

[a] Melting temperature measured by DSC.
[b] Temperature of 10% weight loss measured by TGA.
[c] Temperature of 30% weight loss measured by TGA.
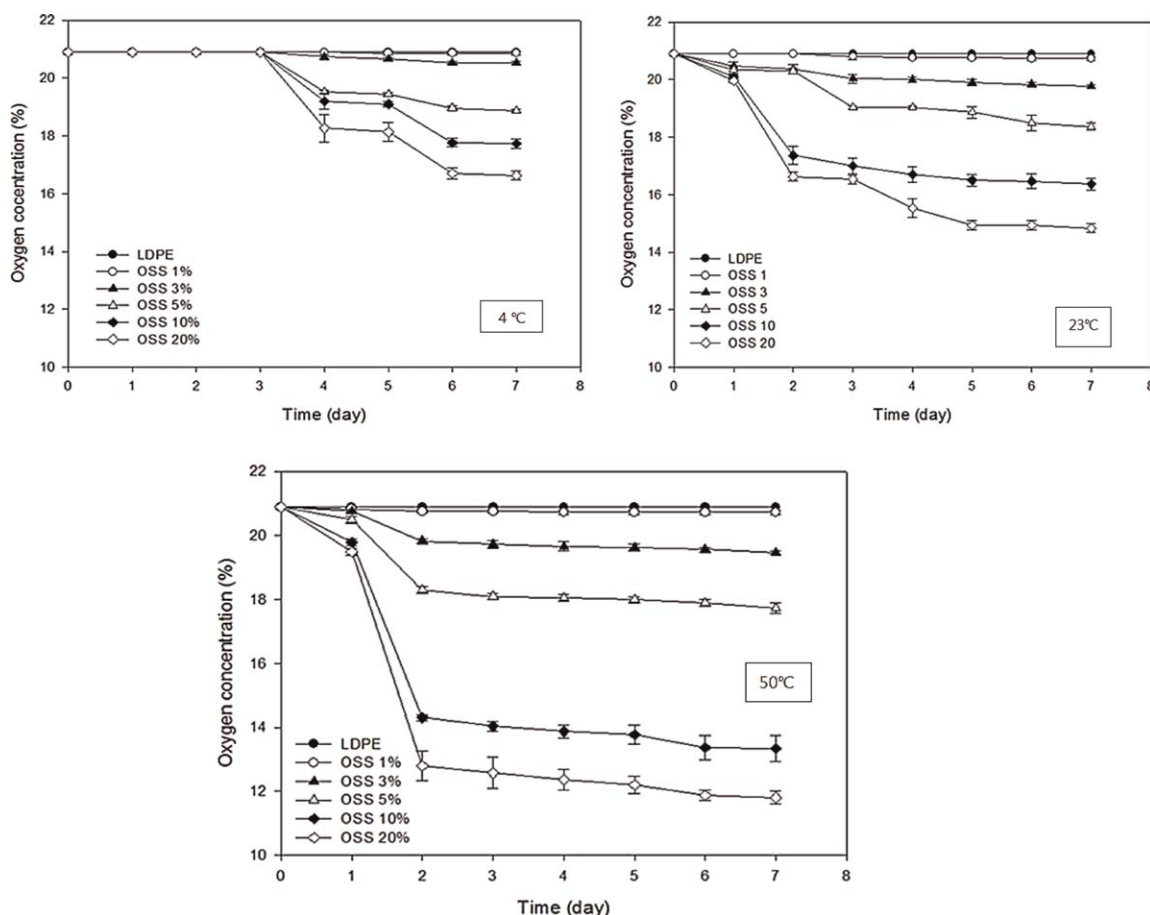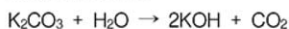[d] Temperature of 50% weight loss measured by TGA.

Figure 7. Oxygen content (%) in the headspace of the films impregnated with different amounts of oxygen scavenging materials at 4 °C, 23 °C, and 50 °C.
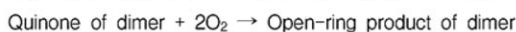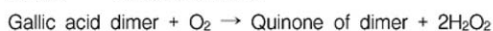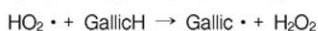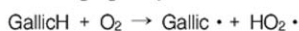
Oxygen Scavenging Capability of Film

The oxygen scavenging capability of the nonmetallic, gallic-acid-based OSS film was significant at 23 °C and at 50 °C as shown in Figure 7. The oxygen scavenging steps involve a reaction among gallic acid, potassium carbonate, and water, as shown in Scheme 1. The oxygen absorption capability (mL/cm^2, 7 days) of

the non-metallic oxygen scavenging films is shown in Table IV. The films containing 10 and 20% of OSS are effective oxygen scavengers at 4, 23, and 50 °C. The pure LDPE (control sample) showed no reduction in oxygen content throughout the entire storage time. The small reduction in headspace oxygen shown for film containing 1, 3, 5% OSS was believed to be due to a physical entrapment of oxygen by LDPE film. RH initiated the oxygen scavenging reaction in the OSS containing film. Water reacts with sodium carbonate and create alkaline environment and gallic acid in alkaline environment start reacting with oxygen present in headspace of glass vial. In this study, the oxygen scavenging capacity from the initial oxygen content and the oxygen content at day 7 was calculated. Based on the results, it is evident that the combinations of the gallic acid and potassium carbonate in LDPE film
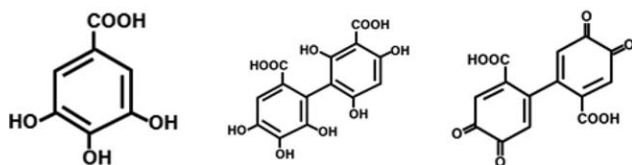
Activation step

$$K_2CO_3 + H_2O \rightarrow 2KOH + CO_2$$

Scavenging step

$$GallicH + O_2 \rightarrow Gallic\cdot + HO_2\cdot$$
$$HO_2\cdot + GallicH \rightarrow Gallic\cdot + H_2O_2$$
$$2Gallic\cdot \rightarrow Gallic\ acid\ dimer$$
$$Gallic\ acid\ dimer + O_2 \rightarrow Quinone\ of\ dimer + 2H_2O_2$$
$$Quinone\ of\ dimer + 2O_2 \rightarrow Open\text{-}ring\ product\ of\ dimer$$

(a) Oxygen scavenging reaction

(b) Gallic acid   **(c) Dimer**   **(d) Quinone**

Scheme 1. Chemical reaction by which gallic acid scavenges oxygen.

Table IV. Oxygen Absorption Capability of Each Oxygen Scavenging Film

Sample	Oxygen absorption capability (mL/cm^2 after 7days)		
	4 °C	23 °C	50 °C
OSS 1%	—	—	—
OSS 3%	0.042	0.130	0.165
OSS 5%	0.234	0.291	0.364
OSS 10%	0.364	0.521	0.870
OSS 20%	0.491	0.709	1.046

has good oxygen scavenging ability. But the scavenging capacity was found to vary with different combinations. Generally, the scavenging capacity of an commercial oxygen scavenging film layer is at least 250 mL O_2/m^2/mil thickness, and it is more often 500 mL O_2/m^2/mil thickness.[37] The film containing 20% OSS had an oxygen scavenging capacity of 0.709 mL/cm^2 over 7 days. The results demonstrate that our developed LDPE film containing a gallic-acid-based OSS could be used as a moisture-activated oxygen-scavenging layer for high moisture containing food including fresh fruits like apple, raw meat and its meat products for active packaging applications.

CONCLUSIONS

In this study, we developed and characterized an LDPE film containing a non-metallic OSS based on gallic acid and potassium carbonate. The SEM results confirmed that the OSS was well incorporated into the film structure. However, agglomerations of the OSS caused a rough film surface, increased water vapor and oxygen permeability, and decreased TS. In the presence of moisture, the oxygen scavenging film can scavenge oxygen effectively. About 20% concentration of non-metallic-based OSS was more effective in terms of oxygen scavenging capacity compared with other sample. The OSS will be useful for actively packaging high-moisture foods like meat and meat products, fresh cut fruits, fruit juices, beer etc. To improve the film's surface morphology, dispersion of the OSS into the LDPE film structure needs to be enhanced, and that will be a focus of our future research work.

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