J. Integrative Natural Sci.
Vol.17, No.4 (2024) pp.157-167
https://doi.org/10.13160/ricns.2024.17.4.157

Comparative Analysis of Functional Hydrogel Lens Containing Gallate and Carotenoid Groups

Seong Hoon Bang and A-Young Sung†

Department of Optometry & Vision Science, Daegu Catholic University

Abstract

The physical and optical properties of functional hydrogel lenses manufactured using Gallate and Carotenoid group materials as additives for hydrogel lenses were analyzed. In order to manufacture a hydrogel lens, a hydrophilic monomer, HEMA (2-hydroxyethyl methacrylate), a cross-linker EGDMA (ethylene glycol dimethacrylate), and a photoinitiator 2H2M (2-hydroxy-2-methylpropyphenone) were used. Additives such as Epigallocatechin gallate, Octyl gallate, and Astaxanthin were added to the basic mixed solution at a ratio, respectively, and photopolymerized with a wavelength of 365 nm for 40 seconds by a cast mold method. The Carotenoid material produced a nano dispersion through the solvent substitution method, and the physical properties of the manufactured contact lens were evaluated by measuring refractive index, water content, spectral transmittance, and contact angle to analyze the changes in physical properties according to the material. Hydrogel contact lenses using the gallate group showed increased wettability and high UV blocking ability compared to base hydrogel lenses. In addition, lenses with epigallocatechin gallate added produced a gradually darker pink color as the amount added increased. Hydrogel lenses using carotenoid nano dispersions showed an increase in both refractive index and water content, and a tendency toward higher wettability. In addition, contact lenses manufactured using an optimized blending ratio of gallate and carotenoid showed increased refractive index, water content, and wettability, as well as antibacterial function and improved UV blocking ability. Hydrogel lenses are manufactured by adding gallate and carotenoid to basic lens material in the ratio can be used as functional medical eye materials with high refractive index, water content, wettability, UV blocking properties, and antibacterial properties.

Keywords : Carotenoid, Gallate, Functional contact lenses, Ophthalmic medical use, UV protection

---

(**Received** September 6, 2024; **Revised** September 11, 2024; **Accepted** October 4, 2024)

†Corresponding author: say123sg@hanmail.net

1. Introduction

Hydrogel lenses used for visual function correction have been released in various designs, materials, and types, and the number of people using them for various purposes such as treatment and beauty as well as vision correction is increasing[1]. Therefore, contact As the use of lenses increases, various studies are being conducted to improve negative aspects such as side effects and discomfort[2]. Additionally, the development of new contact lens materials with various functions such as high wettability, antibacterial properties, UV protection, and oxygen permeability is being developed. Progress is being made actively[3-4]. Nanoparticles can demonstrate the best characteristics and performance with a minimized size and amount, and are a material that is attracting attention in various industrial fields for its various effects such as coloring, antibacterial properties, UV protection, strength, and stability[5]. Astaxanthin is extracted from seaweed and crustaceans, has a red color, and is classified into the carotenoid family. It has a structure similar to β-carotein and has two carboxyl groups[6]. It also strengthens immunity and relieves peripheral fatigue in muscle tissue[7]. Carotenoids are not water soluble, and this property hinders their absorption by the body due to their various advantages, including anti-cancer effects, eye protection, and antioxidant properties. To overcome these problems, nanotechnology can improve the bioavailability and absorbability of lipophilic compounds[8-9]. Solvent displacement is a low energy approach that prevents loss and degradation of heat sensitive substances and separates the organic and aqueous phases. It is a simple method of mixing under magnetic stirring[10]. Therefore, in this study, a solvent substitution method was used to produce nano-dispersions. Gallate, a substance widely used in various fields as an antioxidant material, has an antioxidant effect, so it is more effective than other antioxidants and is heat-stable[11]. Additionally, as an ophthalmic medical material, it increases wettability and has a UV-blocking function[12]. Epigallocatechin gallate, known as the main bioactive ingredient of green tea, is a polyphenol substance and has been shown to have excellent antibacterial effects in removing free radicals and to have antithrombotic, anticancer, antiinflammatory, and antioxidant functions[13]. Antioxidants are a type of compound that has the ability to prevent or slow down the oxidation reaction of other substances[14]. Octyl Gallate is an antioxidant that can be added to foods to inhibit oxidation. In addition, it has been approved as a food additive in many countries and regions, including the European Union (EU) and Australia[15]. In this study, the nano-dispersion produced by the solvent substitution was applied to hydrogel lenses, and gallate and carotenoid materials were used for functional purposes. Used as an additive. In addition, contact lenses were manufactured by photopolymerization and their physical properties were evaluated.

2. Materials and Methods

2.1. Reagents and Materials

To manufacture hydrogel lenses, the main material, the hydrophilic monomer HEMA (2-hydroxyethyl methacrylate), the cross-linking agent EGDMA (Ethylene glycol di-methacrylate), and the photoinitiator 2H2M (2-hydroxy-2-methylpropiophenone) were all products from Sigma-Aldrich. Epigallocatechin gallate, octyl gallate, and astaxanthin used as additives were all from Sigma-Aldrich, and the structural formulas of the additives used in the experiment are shown in **Fig. 1.**

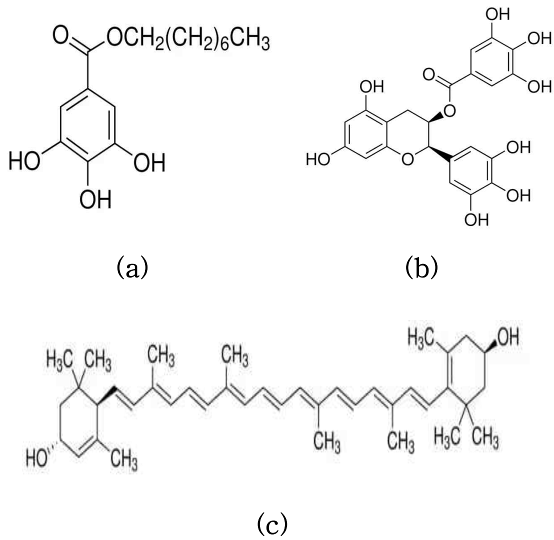


Fig. 1. Chemical structures of additives. (a) Epigallocatechin gallate, (b) Octyl gallate, (c) Astaxanthin.

2.2. Experimental method

In order to produce the hydrogel used in this experiment, hydrophilic monomer HEMA, cross-binding agent EGDMA, and photoinitiator 2H2M were determined as the basic blending ratio. Hydrogel contact lenses manufactured using HEMA were named Ref, and contact lenses were manufactured by adding additives Epigallocate chingallate, Octyl gallate, and Astaxanthin to a mixture of 1%, 5%, and 10% respectively. For the experiment of combining Gallate and Carotenoid, Carotenoid at 1% was fixed and Gallate at 1% and 5% and 10% were added respectively to produce contact lenses. The organic phase was produced by dissolving 1% of Carotenoid in acetone and 0.1% surfactant in deionized water to add Carotenoid to the nano dispersion solution. The organic phase was then added to the aqueous phase in a 1:9 ratio.10) The lenses manufactured by adding Gallate and Carotenoid to the basic hydrogel in the appropriate ratio were named E_1, E_5, E_10, O_1, O_5, O_10, A_1, A_5, and A_10, respectively. The lenses manufactured by combining Gallate and Carotenoid were named Ae_1, Ae_5, Ae_10, Ao_1, Ao_5, and Ao_10, respectively. The contact lenses were manufactured by photopolymerization using a cast mold method with light of 365 nm wavelength for 40 seconds. The compounding ratios of the contact lens samples used in the experiment are shown in **Table 1.**

Table 1. Percent compositions of samples containing Gallate and Carotenoid grouparotenoid group

(Unit : wt%)

Sample	HEMA	EGDMA	2H2M	A[1]	E[2]	O[3]	Total
Ref	98.52	0.99	0.49	-	-	-	100.00
E_1	97.56	0.98	0.49	-	0.98	-	100.00
E_5	93.90	0.94	0.47	-	4.69	-	100.00
E_10	89.69	0.90	0.45	-	8.97	-	100.00
O_1	97.56	0.98	0.49	-	-	0.98	100.00
O_5	93.90	0.94	0.47	-	-	4.69	100.00
O_10	89.69	0.90	0.45	-	-	8.97	100.00
A_1	97.56	0.98	0.49	0.98	-	-	100.00
A_5	93.90	0.94	0.47	4.69	-	-	100.00
A_10	89.69	0.90	0.45	8.97	-	-	100.00
Ae_1	96.62	0.97	0.48	0.97	0.97	-	100.00
Ae_5	93.02	0.93	0.47	0.93	4.65	-	100.00
Ae_10	88.89	0.89	0.44	0.89	8.89	-	100.00
Ao_1	96.62	0.97	0.48	0.97	-	0.97	100.00
Ao_5	93.02	0.93	0.47	0.93	-	4.65	100.00
Ao_10	88.89	0.89	0.44	0.89	-	8.89	100.00

[1]Astaxanthin, [2]Epigallocatechin, [3]Octyl gallate

2.3. Instruments and analysis

The manufactured hydrogel lenses were used in the experiment after being hydrated in 0.9% NaCl physiological saline for 24 hours, respectively, and the refractive index, moisture content, spectral transmittance, contact angle, and antibacterial properties of the lenses were measured, respectively. In order to analyze the optical properties of the manufactured samples, it was measured using Agilent Co. Cary 60 UV-vis, and the spectral transmittance of the UV-B region (280–315 nm), UV-A region (315-380 nm), and visible light region (380-780 nm), which are the criteria for ISO 8599: 1994 (Optics and Optical Instruments of the Spectral and Luminous Transmission), were measured, respectively. In order to measure the refractive index of the prepared samples, an ABBE refractometer (ATAGO NAR IT, JAPAN) was used and measured based on ISO 18369-4:2006. Water content was measured by gravimetric method based on ISO 18369-4:2006 using an electronic scale Ohaus (PAG 214C, USA). In order to measure the contact angle, it was measured using SEO's Phoenix-300 touch instrument. After removing an appropriate amount of moisture from the surface of the lens, a message drop method was used to measure the spreading angle by dropping distilled water on the surface of the lens in a flat state. For the evaluation of antimicrobial activities against S. aureus and E. coli, the cultured microorganisms were compared and analyzed using a dry film (3M Petrifilm) medium. Each 1 mL of the sample hydrated with bacteria in 0.9% NaCl saline solution in the medium for 24 hours was smeared on a dry film and cultured at 35±1°C for 24 hours. SEM measurement was used to confirm the shape of the nanoparticles distributed in the prepared contact lenses, and FE-SEM (JSM-7500F+EDS, Oxford) was used. All experiments were performed by measuring

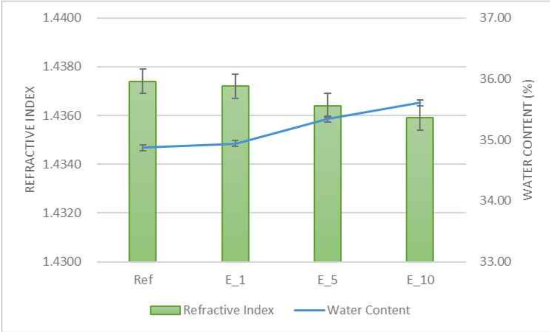
5 times per sample and calculating the average value.

3. Results and Discussions

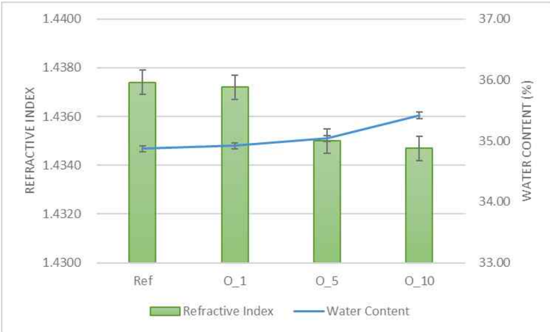
3.1. Characteristics of hydrogel with gallate group

3.1.1. Refractive index and water content

As a result of measuring the refractive index and water content of the manufactured hydrogel contact lens, the basic hydrogel lens (Ref) showed a refractive index of 1.4374 and a water content of 34.87%, respectively. Depending on the addition ratio of epigallocatechin gallate, the refractive index gradually decreased to E_1 1.4372, E_5 1.4364, and E_10 1.4359, and the water content increased to E_1 34.94%, E_5 35.34%, and E_10% 35.61%, showing a tendency for the refractive index and water content to be inversely proportional. Additionally, in case of samples containing octyl gallate, the refractive index values were O_1 1.4372, O_5 1.4350, and O_10 1.4347, respectively. In the case of water content, it was measured as 34.92% for O_1, 35.04% for O_5, and 35.42% for O_10, and the refractive index decreased and water content increased depending on the amount added. The refractive index and water content of each sample are shown in **Fig. 2.**



(a)

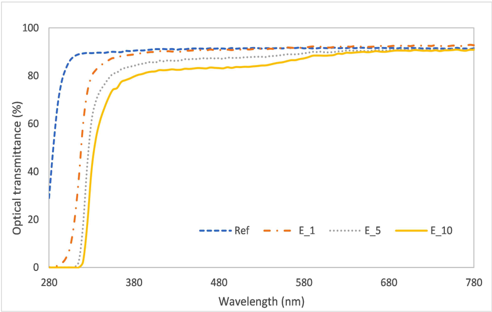


(b)

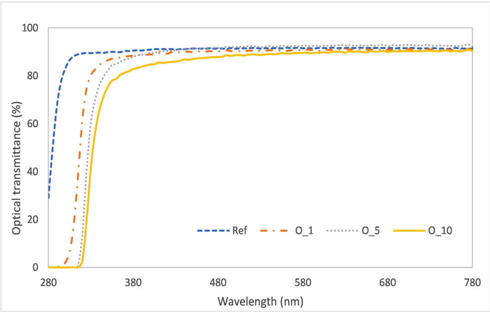
Fig. 2. Refractive index and water content of samp les. (a) Epigallocatechin gallate, (b) Octyl gallate.

3.1.2. Spectral transmittance

As a result of measuring the spectral transmittance of a lens with a gallate group added to the basic hydrogel material, the spectral transmittance of Ref was UV-B, UV-A and Vis. Areas were 59.64%, 82.03%, and 92.73%, and depending on the addition ratio of Epigallocatechin gallate, E_1 was 7.37%, 80.86%, 93.07%, E_5 0.18%, 74.75%, 90.34%, E_10 0.02%, 63.86%, It was measured at 89.86%. When octyl gallate was added, the results were 8.85%, 81.11%, 89.55% for O_1, 0.27%, 70.20%, 89.49% for O_5, and 0.02%, 58.61%, 87.39% for O_10. The UV blocking rate of both gallates improved depending on the amount added, and there



(a)



(b)

Fig. 3. Spectral transmittance of samples. (a) Epigallocatechin gallate, (b) Octyl gallate.

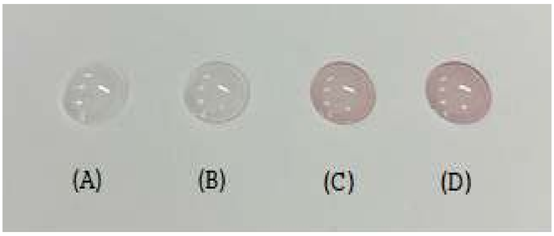


Fig. 4. Produced hydrogel lens of Epigallocatechin Gallate samples. (A):Ref (B):E_1 (C):E_5 (D):E_10.

was no significant difference in the visible light range, so it is believed that they can be used as functional materials for blocking UV rays. The spectral transmittance measurement results of each lens with additives added by ratio are shown in **Fig. 3**, and a photo of the color change of the lens according to the addition ratio of sample E is shown in **Fig. 4**.

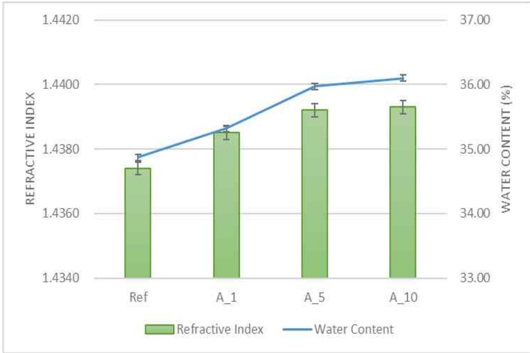


Fig. 5. Refractive index and water content of samples containing Astaxanthin.

3.2. Characteristics of hydrogel with carotenoid group

3.2.1. Refractive index and water content

As a result of measuring the refractive index and water content of hydrogel contact lenses manufactured by adding carotenoid made of nano dispersion using the solvent substitution method, the refractive index increased to A_1 1.4385, A_5 1.4392, and A_10 1.4393 depending on the addition ratio of Astaxanthin, and the moisture water content increased to A_1 35.32%, A_5 35.97%, and A_10% 36.1%, indicating that both refractive index and water content increased. The refractive index and water content of each sample are shown in **Fig. 5.**

3.2.2. Spectral transmittance

As a result of measuring the spectral transmittance of the lens in which the carotenoid nano dispersion was added to the basic hydrogel material, depending on the ratio of Astaxanthin, A_1 was found to be 62.93%, 88.99%, and 92.35% in the UV-B, UV-A and Vis. regions, and A_5

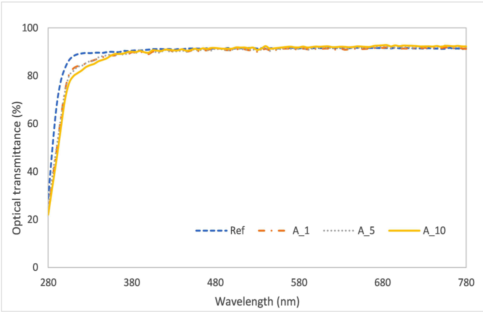


Fig. 6. Spectral transmittance of samples containi ng Astaxanthin

was measured to be 60.49%, 87.99%, 92.05%, and A_10 58.06%, and 87.84%. It is judged that the addition of carotenoid does not significantly affect the spectral transmittance. **Fig. 6** shows the measurement results of the spectral transmittance of the lenses to which the additives are added by ratio.

3.3. Characteristics of hydrogel using a combination of gallate and carotenoid

3.3.1. SEM Analysis

Surface analysis method was used to check the surface condition of the hydrogel lens manufactured using carotenoid nano dispersion by SEM. The image results are shown in **Fig. 7**, and nanoparticles generated on the lens surface were confirmed.

3.3.2. Refractive index and water content

Gallate and carotenoid made from nano dispersion were added to the basic hydrogel mixture solution at a set ratio to manufacture lenses, and their physical properties were compared and analyzed. As a result of measuring the refractive index of the manufactured lens, Ae increased to

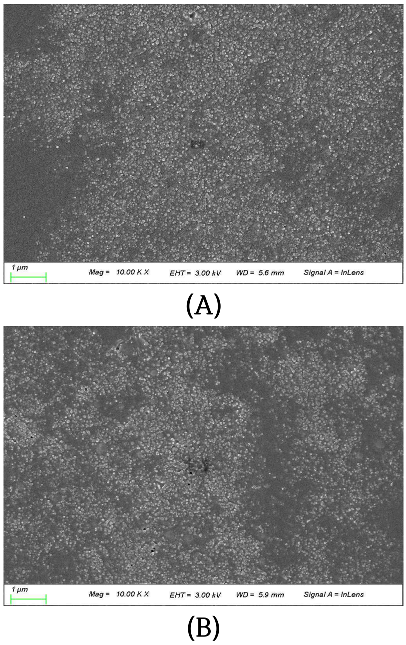


Fig. 7. Surface analysis by SEM image. (A):Ae_5 (B):Ao_5.

the range of 1.4407 to 1.4502 and Ao increased to the range of 1.4415 to 1.4485.

Depending on the amount of additives, water content increased for Ae to 35.46~36.62% and for Ao to 34.9~35.92%. When Epigallocatechin gallate and Octyl gallate were added to carotenoid, there was little difference in physical properties in terms of refractive index. In the case of water content, the combination with the addition of Epigallocatechin gallate was found to be higher. Epigallocatechin gallate contains four epigallocatechin units capable of forming various hydrogen bonds within the molecule. Each unit forms a large number of hydrogen bonds and interacts with water molecules, resulting in a higher water content than octyl gallate. It is judged that The refractive index and water content of each sample are shown in Fig. 8.

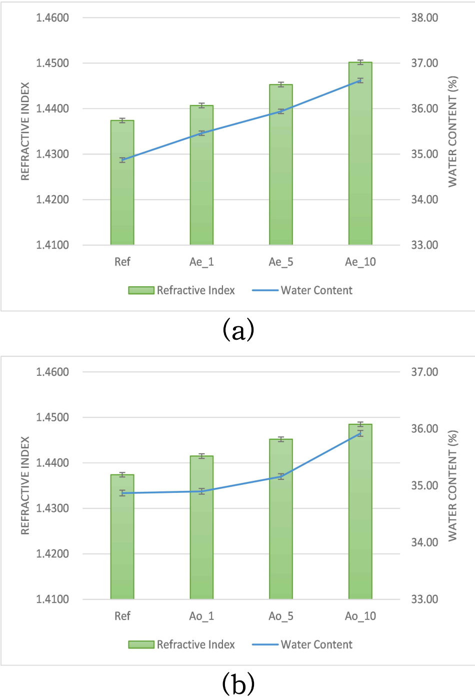


Fig. 8. Refractive index and water content of sam ples. (a) Ae, (b) Ao.

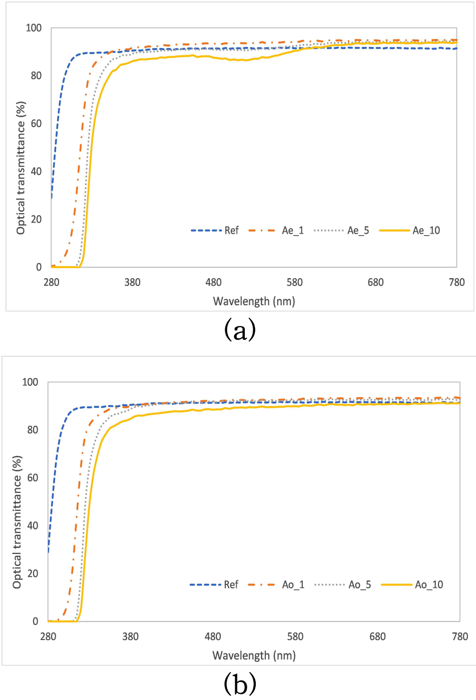


Fig. 9. Spectral transmittance of samples. (a) Ae, (b) Ao.

3.3.3. Spectral transmittance

In case of produced lens by adding gallate and carotenoid together in different ratios, the UV protection rate improved depending on the amount added, and there was no significant difference in the visible light range. According to the medical device standard specifications announced by the Ministry of Food and Drug Safety, the minimum visible light transmittance must be 80% or higher. It was confirmed that the visible light transmittance of all samples was measured to be 80% or higher. The spectral transmittance measurement results of each lens added by ratio are shown in **Fig. 9.**

3.3.4. Contact angle

As a result of measuring the contact angle of the manufactured contact lens, the basic Ref without adding gallate and carotenoid was found to be 64.23°. Ae was found to be 56.63~53.27°, and Ao was found to be 43.86~38.18°. It was confirmed that all samples had lower contact angles than Ref depending on the amount added, showing an increase in wettability. The contact angle results for each lens are shown in **Fig. 10.**

3.3.5. Antimicrobial property

The antibacterial properties of lenses made by adding gallate and carotenoid were evaluated. Ref and sample were used as

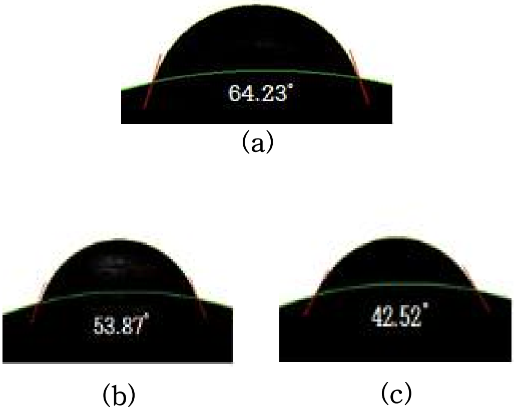


Fig. 10. Contact angle image of samples. (a) Ref, (b) Ae_5, (c) Ao_5.

control and experimental groups, respectively, and the antibacterial properties against Staphylococcus aureus and Escherichia coli were analyzed using the dry film method. 1 mL of the sample that had been hydrated in 0.9% NaCl saline for one week was poured into the dry medium to allow the medium to absorb the sample, and an incubator was used to maintain the temperature at 35°C±1°C. After culturing for about 24 hours, the antibacterial activity against Staphylococcus aureus and Escherichia coli was analyzed. As a result, many microorganisms were identified in Ref, and samples containing gallate and carotenoid showed excellent antibacterial properties against staphylococci and Escherichia coli. It is believed that functional contact lenses with antibacterial properties can be manufactured. The antibacterial results against two types of bacteria are shown in **Fig. 11-12** respectively.

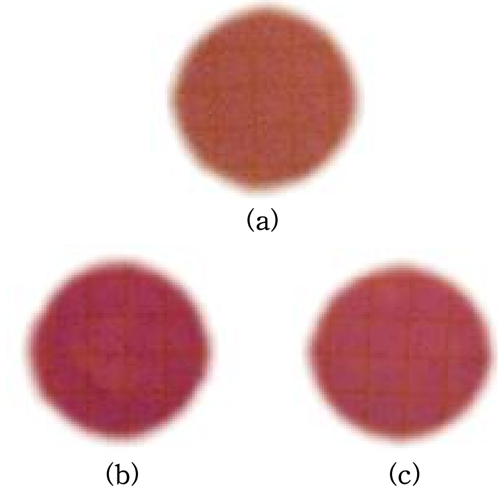


Fig. 11. Antimicrobial property of samples. (a) Ref, (b) Ae_5, (c) Ao_5.

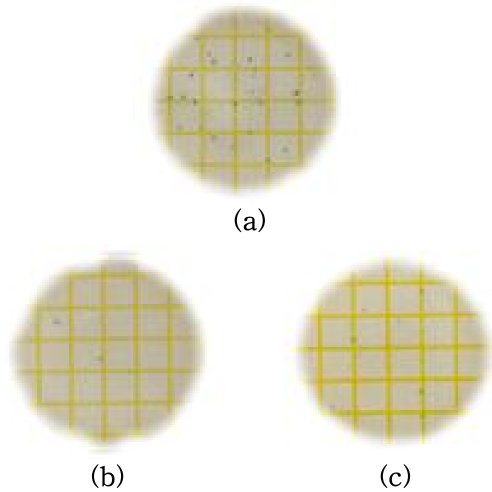


Fig. 12. Antimicrobial property of samples. (a) Ref, (b) Ae_5, (c) Ao_5.

4. Conclusion

In this study, various types of gallate were added in different proportions to manufacture functional hydrogel lenses, and carotenoid was made into a nano dispersion, added in different proportions, then photopolymerized, and the optical and physical properties of the lenses were measured. As a result of using the gallate group as an additive, both Epigallocatechin gallate and Octyl gallate showed an inverse tendency in which water content increased slightly and refractive index decreased slightly depending on the amount added, but no significant change was observed. In addition, it has excellent UV protection and is considered suitable as a material for functional hydrogel lenses. In addition, lenses made by adding epigallocatechin gallate appear pink in color as the amount added increases, so it is believed that they can be used as cosmetic hydrogel lenses. To add carotenoids to hydrogel lenses, a nano dispersion was prepared using a solvent substitution method. And as a result of measuring the optical and physical properties of the contact lens using the nano dispersion, both the refractive index and the water content increased, and the light transmittance was similar to that of the basic hydrogel lens. Therefore, when Astaxanthin is prepared as a nano dispersion, it is judged that it can be used as an additive for a functional hydrogel lens. In addition, after manufacturing a functional hydrogel lens using an optimized mixing ratio of gallate and carotenoid, the refractive index, water content, and wettability all increased compared to the HEMA-based hydrogel lens, which is Ref, in all samples, and the antibacterial function and UV protection were excellent. It is judged that the gallate and carotenoid group materials can be usefully used as a material and manufacturing method for high-functional hydrogel lenses.

References

[1] Young Joo Lee, Seo Yeon Park and Young Sub Eom, “Characteristics of Silicone Hydrogel Contact Lenses Commercially Available in Korea.”, Ann. Optom. Contact Lens, Vol. 21, No. 2, pp. 47-52, 2022.

[2] S. Martin and O. Barr, “Preventing complications in people who wear contact lenses.”, Br. J. Nurs., Vol. 6 No. 11 pp. 614-619, 2014.

[3] Tae Hun Kim, Dong Hyun Kim and A Young Sung, “Optical application of phthalocyanine group for high performance tinted contact lenses.”, Korean Soc. Vis. Sci., Vol. 15, No. 2, pp. 147-156, 2013.

[4] Lee M. J. and Sung A. Y., “Electromagnetic Wave Blocking Effect of Ophthalmological Hydrophilic Lenses Material Containing Indium Tin Oxide Nanoparticles.”, J. Nanosci. Nanotechnol., Vol. 17, No. 10, pp. 7400-7404, 2017.

[5] Seung Kuk Baek, “Clinical Application of Nanotechnology.” Korean J. Otorhinolaryngol. Head Neck Surg., Vol. 54 No. 3, pp. 185-191, 2011.

[6] Naguib and Yousry M A, “Antioxidant activities of astaxanthin and related carotenoids.”, J. Agric. Food Chem., Vol. 48, No. 4, pp. 1150-1154, 2000.

[7] Jin-Kyu Seo1, Jun-Young Park, Beom-Hak Kim, Hyun-Jun Lee, Seong-Gyun Kang

and Han-Sul Yang, “Effects of Astaxanthin on the Oxidative Stability and Quality Characteristics of Emulsified Sausages during Cold Storage.”, J. Agric. Life Sci., Vol. 53, No. 3, pp. 41-49, 2019.
[8] Anton N., Gayet P., Benoit J. P. and Saulnier P., “Nano-emulsions and nano-capsules by the PIT method: an investigation on the role of the temperature cycling on the emulsion phase inversion.”, Int. J. Pharm., Vol. 344, No. 1-2, pp. 44-52, 2007.
[9] Guttoff M., Saberi A. H. and McClements D. J., “Formation of vitamin D nano-emulsion-based delivery systems by spontaneous emulsification: factors affecting particle size and stability.”, Food chem., Vol. 171, pp. 117-122, 2015.
[10] Saberi A. H., Fang Y. and McClements D. J., “Fabrication of vitamin E-enriched nanoemulsions: Factors affecting particle size using spontaneous emulsification.”, J. Colloid Interface sci., Vol. 391, pp. 95-102, 2013.
[11] Park Se-Young and Sung A-Young, “Optophysical Properties of Hydrogel Ophthalmic Lenses Containing Gallate Group.” J. Korean Chem. Soc., Vol. 56, No. 6, pp. 725-730, 2012.
[12] Su-Mi Shin and A-Young Sung, “A Study on Functional Color Contact Lens Materials Including Gallate Group Additives.” Korean J. Vis. Sci., Vol. 25, No. 2, pp. 137-144, 2023
[13] Sang-Mook Lim and Koon-Ja Lee, “Synergistic Antimicorbial Effect of Epigallocatechin-gallate (EGCG) and Polyhexamethylene-biguanide (PHMB) Mixture.” Korean J. Vis. Sci., Vol. 14, No.4, pp. 431-424, 2012.
[14] Sivasankaran U., Vikraman A. E., Thomas D. and Kumar K. G., “Nanomolar Level Determination of Octyl Gallate in Fats and Oils.” Food Anal. Methods, Vol. 9, pp. 2115–2123, 2016.
[15] Wang Q., Zhang Y., and Li H., “Octyl gallate: An antioxidant demonstrating selective and sensitive fluorescent property.”, Food Chem., Vol. 219, pp. 268-273, 2017.