Sustainable active packaging for the EU green transition: HEC/vitamin C antioxidant films enhancing food safety and reducing waste

ABSTRACT

The growing demand for sustainable food packaging is driven not only by consumer awareness of food safety but also by international strategies such as national development plans and European Union regulations, which emphasize the replacement of conventional plastics with bio-based and functional alternatives. In this context, active antioxidant packaging plays a critical role in extending food shelf life, reducing waste, and ensuring safer products. Hydroxyethyl cellulose (HEC) was selected as the matrix material due to its good film-forming properties, biodegradability, biocompatibility, odorlessness, and non-toxicity or low toxicity, while vitamin C (ascorbic acid), a natural antioxidant, was incorporated as the active agent. HEC solution was prepared by dissolving HEC powder in distilled water, and vitamin C was incorporated. The resulting mixtures were homogenized and cast onto glass plates, followed by drying to produce uniform films. The obtained films were systematically characterized in terms of contact angle (wettability), and antioxidant capacity. In addition, their surface performance was evaluated through IGT F1 flexographic test printing trials to assess practical applicability. The results demonstrated that the incorporation of vitamin C enhanced the antioxidant functionality of HEC films, while altering their barrier and surface properties depending on the concentration used. These findings suggest that HEC-based films containing vitamin C have significant potential as active packaging materials that meet both sustainability goals and food safety requirements.

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Introduction

Plastic packaging is widely preferred in the food industry due to its low cost, ease of production, good barrier properties, and ability to protect products against both chemical degradation and physical impacts encountered during the distribution process (Chen et al., 2024; Haghighi-Manesh & Azizi, 2017). However, the widespread use of these materials and their slow biodegradation pose various risks to both environmental pollution and health. The rise of single-use plastic packaging is seen as one of the primary causes of this problem (Rojas et al., 2021; Velásquez et al., 2021).

In recent years, the environmental problems caused by plastic waste have attracted public attention and led to increased consumer demand for unprocessed and additive-free products (Gao et al., 2023). Consequently, the need for packaging materials is both diversifying and increasing. In this regard, the European Union's new Packaging and Packaging Waste Regulation (PPWR) mandates critical steps in the food packaging sector within the framework of circular economy and sustainability goals. The regulation aims to ensure that all packaging is recyclable by 2030, that plastic packaging contains a certain percentage of recycled materials, and that reusable systems are expanded.

This regulation aims to minimize the environmental impact of packaging and encourage the development of environmentally friendly materials produced from sustainable sources (Mohammadi et al., 2019; Shankar, Wang & Rhim, 2019).

Furthermore, in recent years, there has been a notable increase in the use of films made from natural polymers due to their non-toxicity and biocompatibility (Si et al., 2022; Kanatt & Makwana, 2020).

The primary function of food packaging is to protect products against air, moisture, dust, light, and chemical and microbiological contamination. Another important function of packaging materials is to extend the shelf life of products. However, most traditional packaging is made from non-biodegradable plastics. The inability to eliminate these plastics has led to increasing calls from consumers and environmentalists to reduce unnecessary plastic use in packaging (Velásquez et al., 2021; Arrieta et al., 2019).

Biodegradable polymers are naturally degraded by bacteria, fungi, and other microorganisms, converting biomass into water and carbon dioxide. They are typically produced using plant resources or other renewable materials. In recent years, these polymers have been intensively researched in various fields due to their potential to reduce environmental pollution and alleviate waste management problems (Brockgreitens & Abbas, 2016).

Antioxidant and antimicrobial active packaging refer to methods that aim to limit oxidation and microbial activity to preserve the freshness and quality of foods. These types of packaging materials are prepared using either natural or synthetic ingredients, thereby delaying the oxidation of oils and inhibiting the growth of microorganisms, thus extending the product's shelf life. In antimicrobial packaging, active ingredients are released into the food environment to inhibit the growth of bacteria, yeast, and mold. In contrast, in antioxidant systems, antioxidants are incorporated directly into the packaging material and neutralize free radicals, slowing the oxidative deterioration of oils in food. Examples include vitamin E, ascorbic acid, and plant polyphenols (Zende, Ghase & Jamdar, 2025; Choe & Min, 2006). Active packaging ensures longer shelf life while ensuring fewer additives and preservatives in food formulations, greater flavor retention, and higher food quality. Antioxidant controlled release generally provides long-term food protection against lipid oxidation through the continuous or gradual release of antioxidants, particularly at the food surface (Yam, Schaich & Obinata, 2008; Jamshidian, Tehrany & Desobry, 2013).

HEC is used as a thickener, preservative or binder, suspension, and colloid stabilizer in cosmetics, biomedical, pharmaceuticals and coatings and food applications and many more due to its biodegradable, odorless and non-toxic properties and is approved by the U.S. Food and Drug Administration (FDA) (Noreen et al., 2020; El Fawal et al., 2020; Yang & Li, 2018). HEC has poor toughness and load-carrying properties, so combining it with other materials makes it more useful (Cen et al., 2023). Studies in the literature have investigated the use of nanoparticulate HEC as a body scaffold material in biomedical engineering (Zulkifli et al., 2017), its use in combination with hyaluronic acid for wound dressing (Luo et al., 2018), the enhancement of UV resistance and mechanical properties through nanofiber reinforcement (Huang et al., 2021), and the enhancement of solvent resistance and load-carrying capacity, as well as UV protection properties, of HEC films through the addition of zinc oxide and carboxymethyl chitosan (Cen et al., 2023).

HEC is expected to provide high UV protection in out-door conditions, but it lacks both mechanical and UV protection. Experiments are underway with CNF, CNC, and inorganic materials to address these weaknesses (Lu et al., 2020). Vitamin C, also known as ascorbic acid, is a naturally occurring plant-based compound with high polarity and high antioxidant and antimicrobial activity. It is a water-based compound found in fruits such as mangoes, oranges, lemons, and blackberries, as well as various vegetables (Zou et al., 2016). Vitamin C has been incorporated into whey protein (Min & Krochta, 2007) and alginate (De'Nobili et al., 2016) based films, along with aloe vera coating gel (Sogvar, Saba & Emamifar, 2016), to provide food protection.

In this study, an active antioxidant packaging film will be produced using hydroxyethyl cellulose (HEC). Vitamin C (ascorbic acid), a natural antioxidant, will be added as the active ingredient. The films will be subjected to the necessary characterization and antioxidant tests. Additionally, to evaluate printability parameters, prints will be made using an IGT F1 flexo printability test press, and relevant parameters will be evaluated.

Experimental

Materials

Hydroxyethyl cellulose (HEC) powder was purchased from Sigma-Aldrich (USA), while glycerol used as a plasticizer and L-ascorbic acid as an antioxidant were obtained from Merck (Germany). Polystyrene petri dishes were sourced from Greiner Bio-One (Germany).

Process blue ink was (Siegwerk, Germany) employed for printability assessments. The antioxidant reagent 2,2-diphenyl-1-picrylhydrazyl (DPPH) and analytical-grade methanol were both procured from Sigma-Aldrich (USA). Adhesion tests utilized Tesa tape (Tesa SE, Germany).

All reagents and materials were of analytical grade or higher to ensure experimental reliability and reproducibility.

Methods

Active packaging films were prepared following the method described by Chen et al. (2024) with modifications. A 2% (w/v) HEC solution was prepared by dissolving 2.0 g of HEC powder in 100 mL of distilled water under continuous magnetic stirring at 50°C until complete homogenization was achieved. Glycerol was added as a plasticizer at 30% (w/w) relative to the weight of HEC (i.e., 0.6 g glycerol per 2.0 g HEC). Subsequently, ascorbic acid was incorporated into the formulation at 5% (w/w) of the total solid content (HEC + glycerol + ascorbic acid).

The resulting film-forming solution was cast onto polystyrene petri dishes and dried at 40°C for 24 h in a ventilated oven. Dried films were then carefully peeled off and conditioned at 25 \pm 2°C and 50 \pm 5% relative humidity for 48 h prior to characterization.

Color characteristics of the films were evaluated using an X-Rite eXact portable spectrophotometer, following the ISO 13655:2017 standard. The device operated with a 0/45° geometry and polarization filter, using a 2° standard observer angle and D50 illumination. Measurements were performed across the visible spectrum (400–700 nm). A white background with L^* = 95.98, a^* = -1.36, and b^* = 1.63 was used as the reference. Each sample was measured four times, and the total color difference (ΔE_{oo}) and whiteness index (*WI*) were calculated to evaluate optical quality.

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T \frac{\Delta C' \Delta H'}{k_C S_C k_H S_H}}$$
(1)

$$WI = 100 - \sqrt{(100 - L)^2 + a^2 + b^2}$$
 (2)

Color differences between the films and the reference background were determined using the CIEDE2000 formula (ΔE_{oo}), in accordance with the ISO 11664-6:2014 standard. Calculations were based on the mean values from five independent measurements per sample. In this method, ΔL^* , Δa^* , and Δb^* represent the differences in lightness, red—green, and yellow—blue coordinates, respectively, between the sample and the reference. The L^* axis denotes lightness (ranging from black to white), the a^* axis spans from green (negative values) to red (positive values), and the b^* axis ranges from blue (negative) to yellow (positive).

The ΔE_{oo} formula accounts for perceptual non-uniformities by incorporating ΔL^* (lightness difference), ΔC^* (chroma difference), and ΔH^* (hue difference), along with an interactive rotation term (*RT*) that

addresses the interplay between chroma and hue in the blue region. Weighting functions SL, SC, and SH are applied to adjust the influence of lightness, chroma, and hue components based on the color's position in CIE $L^*a^*b^*$ space. Parametric factors kL, kC, and kH, typically set to 1 under standard conditions, account for variations due to observation conditions such as texture, background, and sample geometry.

Gloss properties of the films were measured using a BYK-Gardner micro-gloss meter at a fixed incident angle of 75°, following the ISO 8254-1:2009 standard. All measurements were conducted under consistent environmental conditions to ensure accuracy and reproducibility.

Surface wettability was evaluated via static contact angle analysis using a Pocket Goniometer (Model PG-X+, Version 3.4, FIBRO Systems AB, Sweden). Deionized water droplets were carefully dispensed onto the film surfaces, and the contact angles were recorded using a CCD video camera system. Surface free energy (SFE) was subsequently calculated in accordance with ASTM D5946, based on the measured water contact angles. Surface morphology was examined using a Troika SurfaceCAM HD 3D microscope (Troika Systems, UK).

The printability of the films was assessed using an IGT F1 flexographic test printing system (IGT Testing Systems, Netherlands), print settings are printing force of 250 N/M and printing speed of 0.30 m/s. The anilox cell frequency is 150 lpi. Siegwerk process blue ink as the printing ink. Printed surfaces were evaluated for color uniformity using an X-Rite eXact spectrophotometer (X-Rite, USA), and print adhesion was examined by applying and removing Tesa tape (Tesa SE, Germany) in accordance with standard tape test procedures.

The integrity of the printed layer after tape removal was used to qualitatively assess ink adhesion performance.

The antioxidant activity of the active films was determined by measuring their DPPH (2,2-diphenyl-1-pic-rylhydrazyl) radical scavenging capacity, following the method described by Doh, Dunno & Whiteside (2020). The antioxidant potential of the active films was assessed using the 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging assay.

Film samples (20 mg) were immersed in 5 mL of methanol and incubated at 25°C for 12 h to extract antioxidant compounds. Following incubation, 0.2 mL of the film extract was mixed with 2.0 mL of freshly prepared DPPH solution (0.025 g/L in methanol). The mixture was then incubated in the dark at 25°C for 30 minutes to allow for the reaction between antioxidants and DPPH radicals. The absorbance of the reaction mixture was subsequently measured at 517 nm using a UV—visible spectrophotometer (Shimadzu, Japan).

The DPPH radical scavenging activity (RSA) was calculated using the following equation:

DPPH scavenging effect (%) =
$$\frac{A_{DPPH} - A_{extract}}{A_{DPPH}} \times 100$$
 (3)

where, and are the absorbance values of the solution of DPPH and the sample extracts, respectively. The experiment was carried out in triplicate for each film sample, and the results were expressed as mean values ± standard deviation.

Results and Discussion

Hydroxyethyl cellulose (HEC)-based films, both with and without vitamin C, were successfully fabricated, exhibiting uniform, continuous, and defect-free surfaces. The films were subsequently characterized for a comprehensive set of properties, including optical attributes (color, gloss, whiteness index), transparency, water contact angle, antioxidant activity, and surface morphology.

These analyses confirmed that the incorporation of vitamin C did not compromise the structural integrity of the films, while allowing evaluation of the functional effects of the bioactive compound on both physicochemical and optical properties. The successful production and systematic characterization of these films provide a solid foundation for investigating their potential applications in active packaging and other functional biomaterial fields.

The incorporation of vitamin C into the hydroxyethyl cellulose–glycerol film formulation led to a noticeable modification in the optical properties, as evidenced by the CIE color parameters (Table 1). While the control film (without vitamin C) exhibited minimal changes in color, the vitamin C-loaded film displayed a yellowish hue when measured against a white background.

This phenomenon can be attributed to the intrinsic optical characteristics of vitamin C and its distribution within the polymeric matrix. Similar observations have been reported in the literature, where the addition of bioactive compounds, particularly vitamins and phenolic antioxidants, induced slight yellowing or enhanced color intensity in biopolymer-based films (Periyasamy, Asrafali & Lee, 2025).

In contrast, the hydroxyethyl cellulose-glycerol film without vitamin C maintained high color stability, which is consistent with previous findings indicating that polysaccharide—plasticizer matrices generally provide high transparency and minimal coloration unless bioactive additives are incorporated (Sothornvit & Krochta, 2000; Rhim, Park & Ha, 2013).

Therefore, the yellowish tone observed in the vitamin C-loaded film can be considered consistent with the literature and is mainly attributed to the inherent color properties of vitamin C and its influence on the optical behavior of the polymeric matrix.

Color differences (ΔE_{oo}) were calculated using the CIE ΔE_{oo} formula with the white paper as the reference. The HEC film placed on white paper exhibited a minimal color difference of 1.77, indicating negligible visual alteration relative to the substrate. In contrast, the HEC film containing vitamin C showed a markedly higher color difference of 28.01, reflecting a pronounced yellowish shift. This substantial ΔE_{oo} value indicates that vitamin C significantly affects the visual appearance of the film when applied to a white background.

These results are consistent with previous studies reporting that the incorporation of bioactive compounds, such as vitamins or phenolic antioxidants, into polysaccharide-based films can induce notable color changes, often leading to yellowing or other perceptible chromatic shifts (Pan et al., 2024). The contrast between the minimal ΔE_{oo} of the HEC film without vitamin C and the pronounced ΔE_{oo} of the vitamin C-loaded film underscores the strong influence of functional additives on the optical and colorimetric properties of biodegradable films.

Although the vitamin C-loaded HEC film exhibited a noticeable yellowish hue when placed on a white substrate, the whiteness index (WI) calculated according to the ASTM E313 standard increased (Table 1). This phenomenon can be attributed to the contribution of the b^* (yellow-blue) component in the ASTM E313 formula, which positively influences the WI value.

Consequently, even though the film appears yellow to the naked eye, the enhanced b^* value results in a higher calculated whiteness index, reflecting a measurable change in optical properties.

Table 1Colorimetric and optical parameters of unprinted hydroxyethyl cellulose–glycerol films with and without vitamin C

	L*	a*	b*	ΔE _{oo}	Gloss	WI
Base white paper	90.45	3.05	-11.96	Ref.	5.2	15.61
Unprinted HEC film	87.67	3.12	-11.71	1.77	11.2	17.29
Unprinted vitamin C-loaded HEC film	84.27	2.14	27.18	28.01	9.6	31.48

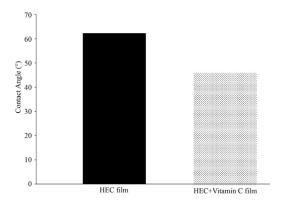
Similar observations have been reported in the literature, where the addition of vitamins or phenolic compounds to biopolymer-based films caused a slight yellowing yet led to an increase in the whiteness index (Rhim, Park & Ha, 2013). These findings indicate that vitamin C affects not only the visible color of HEC films but also the whiteness index as quantified by standard colorimetric methods.

The gloss values of the films were also evaluated to assess their surface optical properties. The white paper exhibited a gloss of 5.2, whereas the HEC film on a white substrate demonstrated a higher gloss of 11.2, indicating an increase in surface reflectivity due to the polymer matrix. Interestingly, the incorporation of vitamin C slightly reduced the gloss to 9.6 compared to the HEC film without vitamin C, suggesting that the presence of the bioactive compound modifies the surface characteristics of the film.

These findings are consistent with literature reports indicating that the addition of functional additives, such as vitamins, phenolics, or other bioactive compounds, can influence the surface gloss of biopolymer films by altering the film's microstructure and light scattering properties (Sothornvit & Krochta, 2000). The comparison among the samples demonstrates that while the HEC matrix inherently increases surface reflectivity relative to the base paper, the incorporation of vitamin C slightly decreases gloss, likely due to changes in surface morphology or light absorption characteristics.

The water contact angles (*WCA*) of the films were measured to assess their surface wettability (Figure 1). The HEC film exhibited a *WCA* of 62.3°, indicating a moderately hydrophilic surface.

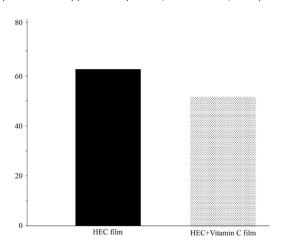
Upon incorporation of vitamin C, the WCA decreased to 46.3°, demonstrating an increase in surface hydrophilicity. This decrease can be attributed to the introduction of polar hydroxyl groups from vitamin C, which enhance hydrogen bonding interactions with water molecules, thereby improving the film's wettability.



» Figure 1: Contact angle of HEC film and vitamin C loaded HEC film

These findings agree with previous studies reporting that the addition of hydrophilic bioactive compounds, such as vitamins or phenolic antioxidants, can significantly reduce the contact angle of polysaccharide-based films, enhancing their hydrophilic character (Cui et al., 2023). The comparison between the two films highlights the pronounced effect of vitamin C on the surface properties of HEC films, suggesting potential implications for applications requiring improved water interaction or controlled wettability.

The antioxidant activity of the films was evaluated using the DPPH radical scavenging assay to determine the functional contribution of the incorporated bioactive compound (Figure 2). The pristine HEC film exhibited a negligible scavenging activity of 2%, consistent with the inherent lack of antioxidant groups in polysaccharide matrices. In contrast, the HEC film containing vitamin C demonstrated a markedly enhanced radical scavenging activity of 58%, confirming the effective integration and functionality of the bioactive molecule within the polymer network. The substantial increase in DPPH inhibition highlights the ability of vitamin C to provide potent antioxidant properties while maintaining the structural integrity of the film. These findings align with previous studies reporting that the addition of hydrophilic antioxidants such as vitamins or phenolic compounds can significantly elevate the radical scavenging capacity of polysaccharide-based films, transforming chemically inert matrices into functional materials suitable for active packaging and food preservation applications (Pereira, Lonni & Mali, 2022).

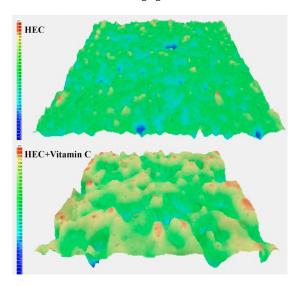


» Figure 2: The antioxidant activity of the HEC film and vitamin C loaded film

Surface morphology of the films was evaluated using a Troika AniCAM HD+ 3D scan microscope. The microscope images given in Figure 3.

The neat HEC film exhibited a smooth and uniform surface with a low roughness value of approximately 0.2 μm , indicating a homogeneous polymeric matrix. In contrast, the incorporation of vitamin C clearly modified the surface microstructure.

The vitamin C-loaded HEC film displayed discernible crystalline particulates that were homogeneously distributed across the surface, with localized roughness values reaching up to $^{\sim}1~\mu m$. This morphological alteration indicates that vitamin C acted not only as a functional additive but also as a structuring agent within the film matrix.



» Figure 3: Surface topography of HEC film and vitamin C-loaded HEC film

These findings are in close agreement with the contact angle and gloss measurements. The vitamin C-loaded film exhibited a lower water contact angle (46.3°) compared to the neat HEC film (62.3°), which can be attributed to the increased hydrophilicity and enhanced surface heterogeneity introduced by vitamin C. Likewise, the reduction in surface gloss of the vitamin C-containing film is consistent with the higher micro-scale roughness observed in the topographical analysis.

Together, these complementary results confirm that the incorporation of vitamin C significantly influences both the physical and surface-related properties of HEC films.

The observed trends are consistent with previous reports in the literature, which have demonstrated that the addition of bioactive compounds or antioxidants into biopolymer matrices increases surface roughness, alters wettability, and reduces optical gloss due to microstructural heterogeneity (Mikus & Galus, 2025).

Overall, the combination of microscopy, wettability, and optical analyses indicates that vitamin C not only enhanced the antioxidant functionality of HEC films but also induced pronounced changes in surface topography in a manner fully consistent with earlier findings on active packaging materials.

Flexographic printability of the prepared films was evaluated using an IGT F1 printability tester. Both the neat HEC film and the vitamin C-loaded HEC film provided defect-free surfaces for solid area printing, indicating that the substrate morphology did not hinder ink transfer or coverage. The colorimetric parameters (CIE $L^*a^*b^*$), gloss, and ΔE_{oo} values of the obtained prints are summarized in Table 2.

A clear shift toward the yellow region of the color space was observed for the vitamin C-containing film, which can be ascribed to the intrinsic coloration and distribution of vitamin C within the polymeric matrix.

This finding implies that when such films are intended for use as printable packaging substrates, appropriate color management and ink formulation adjustments are necessary to achieve target shades and minimize deviations in brand color consistency. The adhesion of the printed flexographic ink was evaluated using the standardized tape (cross-hatch) test, which is a widely accepted and ISO/ASTM-standardized qualitative method for assessing print and coating adhesion on flexible polymeric substrates.

According to ASTM D3359 and ISO 2409, the absence of any visible ink residue transferred onto the tape is classified as the highest adhesion rating (5B) and is considered evidence of excellent anchorage between the ink layer and the substrate.

This outcome was indeed observed in our study, indicating that no delamination or cohesive failure occurred and confirming that the incorporation of vitamin C did not adversely affect the printability or adhesion performance. The tape test was selected because it is routinely employed in both industrial practice and academic research as a fast, reproducible, and directly comparable screening method for printed or coated packaging films, particularly in flexographic applications.

Table 2Colorimetric and optical parameters of flexographic printed hydroxyethyl cellulose–glycerol films with and without vitamin C

	L*	a*	b*	ΔE _{oo}	Gloss
Printed base white paper	85.30	-7.97	-18.46	Ref.	8.9
Printed HEC film	82.81	-8.40	-18.94	1.70	14.7
Printed vitamin C-loaded HEC film	71.62	-6.98	-7.63	11.72	11.1

Conclusions

In this study, HEC-based films incorporating vitamin C were successfully fabricated and comprehensively characterized, revealing a unique combination of functional and processable properties. The incorporation of vitamin C not only enhanced the antioxidant activity of the films but also conferred notable oxygen-scavenging capacity, a feature of relevance for active packaging applications aimed at extending shelf life and preserving product quality. Optical, surface, and wettability analyses demonstrated that the films maintained structural integrity and uniformity, while subtle modifications in color, gloss, and surface roughness reflected the controlled incorporation of the bioactive compound.

Importantly, flexographic printability tests confirmed defect-free ink transfer and excellent adhesion, highlighting the compatibility of these functional films with conventional packaging processes.

The synergy between bioactivity, oxygen scavenging, and industrially relevant printability underscores the originality of this work, providing a rare example of a biopolymer-based material that combines active functionality with practical application potential.

Overall, this study highlights the promise of vitamin C-loaded HEC films as sustainable, functional, and printable substrates for active packaging, bridging the gap between laboratory-scale material innovation and industrial implementation. The demonstrated oxygen scavenging, and antioxidant functionalities position these films as compelling candidates for next-generation packaging solutions that actively contribute to food preservation and quality maintenance.

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