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Bluberry extract: application in smart packaging based no poly(vinyl)alcohol and gelatin for fish products

Bárbara Teixeira Gomes
Doctor Scientiae

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Thesis submitted to the Food Science and Technology Graduate Program of the Universidade Federal de Viçosa in partial fulfillment of the requirements for the degree of *Doctor Scientiae*.

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Assent:

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Adviser

I dedicate this doctoral thesis to my parents, who guided me throughout my life, always prioritizing my education. They allowed me to get this far, they gave me support and backing, and thanks to them I have achieved yet another victory. I am sure that they left for a better place, calm, carefree and proud, because knowing me, they knew that I would be able to complete this stage, despite all the difficulties.

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ABSTRACT

GOMES, Bárbara Teixeira, D.Sc., Universidade Federal de Viçosa, February, 2025. **Bluberry extract: application in smart packaging based no poly(vinyl)alcohol and gelatin for fish products**. Adviser: Nilda de Fatima Ferreira Soares. Co-advisers: Taila Veloso de Oliveira and Paulo Cesar Stringheta.

Blueberry extract has a high anthocyanin content, capable of changing color depending on the pH variation of the medium, and is promising for incorporation into polymeric matrix for the production of smart packaging. In this context, this work aimed to investigate the incorporation of three blueberry extracts, crude extract (EB), phenolic extract (EF) and anthocyanin extract (EFA) in films based on poly (vinyl alcohol) PVA and gelatin, focusing on the development of color indicators for monitoring fish quality. Specifically, we sought to evaluate the in vitro colorimetric behavior (volatile acid vapors - acetic acid, and basic vapors - ammonia) of blueberry extracts and films produced by the casting method (film with crude extract (FB), film with anthocyanin extract (FA) and film with phenolic extract (FF)). The sensitivity and color reversibility of the films and their stability over time (30 minutes) in the presence and absence of oxygen at 25 ± 2 ºC were evaluated. The mechanical, physical, chemical, thermal and topographic properties of the films were analyzed. The capacity and effectiveness of FB and FA to monitor the quality of fresh tilapia fillets and gray shrimp stored under refrigeration (6.5 °C) for 7 days were also explored. High levels of phenolic compounds and anthocyanins were observed in EB and EFA, corroborating the colorimetric results for FA, which showed the greatest overall color difference, changing completely from a greenish to purple color in the presence of acid vapors. FB, on the other hand, exhibited differences in hue values (h°), changing completely from a purple to greenish color and was more sensitive to basic vapors. Blueberry extracts increased the thermal stability of the PVA and gelatin polymer blend. The gelatin films (FG) were more rigid (higher Young's Modulus), while PVA contributed to increase the tensile strength and elongation capacity of the polymer blend with gelatin. The isolation of anthocyanin promoted a plasticizing effect, in which FA was the most elastic. Regarding topography, FB, FF and FA exhibited a more heterogeneous surface than the PVA (FPVA) and gelatin control film, due to the incorporation of blueberry extracts that reduced the interaction between PVA and gelatin. The incorporation of EB and EFA hindered the interaction between PVA and gelatin, decreasing the moisture content of FB and FA. The solubility of the polymer blend was reduced in relation to FG and FPVA, showing itself favorable for application in food. FF presented

the lowest solubility in water, which can be attributed to its less hydrophilic nature in relation to FB and FA, since it does not contain anthocyanins. When applied to smart packaging, the FA indicators changed from green to brown, and the FB from purple to green or brown, in response to the degradation of shrimp fillets and fish, indicating deterioration of the shrimp and tilapia fillets, respectively, after 7 days of storage. For shrimp, the FA and FB indicators changed color concomitantly with the increase in the pH (7.93) of the food, this value being considered unfit for consumption according to Normative Instruction No. 23/19, while for fish fillets, the pH values (6.83) were above those permitted by Ordinance No. 1255/62 of RISPOA. The smart and sustainable FB indicator was more stable during storage, sensitive and irreversible to color change, in addition to being simpler and faster to produce, as it does not require the purification step. Therefore, FB is recommended as a colorimetric indicator for monitoring and controlling fish quality.

Keywords: natural extract; blueberry; colorimetric indicator film; poly (vinyl alcohol); gelatin; smart packaging

RESUMO

GOMES, Bárbara Teixeira, D.Sc., Universidade Federal de Viçosa, fevereiro de 2025. **Extrato de Mirtilo Bruto e Purificado: aplicação em embalagens inteligentes a base de poli(vinil)álcool e gelatina para monitoramento de pescados**. Orientadora: Nilda de Fatima Ferreira Soares. Coorientadores: Taila Veloso de Oliveira e Paulo Cesar Stringheta.

O extrato de mirtilo possui alto teor de antocianinas, capazes de alterar de cor em função da variação de pH do meio, sendo promissor para ser incorporado em matriz polimérica para produção de embalagem inteligente. Nesse contexto, este trabalho teve como objetivo investigar a incorporação de três extratos de mirtilo, o extrato bruto (EB), o extrato fenólico (EF) e o extrato de antocianinas (EFA) em filmes à base de poli (vinil álcool) PVA e gelatina, com foco no desenvolvimento de indicadores de cor para monitoramento da qualidade de pescados. Especificamente, buscou-se avaliar o comportamento colorimétrico in vitro (vapores voláteis ácido - ácido acético, e básico - amônia) dos extratos de mirtilo e dos filmes produzidos pelo método de *casting* (filme com extrato bruto (FB), filme com extrato de antocianinas (FA) e filme com extrato fenólico (FF)). Avaliou-se a sensibilidade e reversibilidade de cor dos filmes e sua estabilidade ao longo do tempo (30 minutos), na presença e ausência de oxigênio, a 25 ± 2 ºC. Analisou-se as propriedades mecânicas, físicas, químicas, térmicas e topográficas dos filmes. Também foi explorado a capacidade e a eficácia dos FB e FA de monitorar a qualidade do filé de Tilápia e do camarão cinza, frescos e armazenados sob refrigeração (6.5 °C) por 7 dias. Foi observado altos níveis de compostos fenólicos e antocianinas no EB e EFA, corroborando com os resultados colorimétricos para o FA, que apresentou a maior diferença global de cor, mudando completamente de uma cor esverdeada para roxa na presença de vapores ácidos. Já o FB exibiu diferenças nos valores de tonalidade (h°), mudando completamente de uma cor roxa para esverdeada e foi mais sensível aos vapores básicos. Os extratos de mirtilo aumentaram a estabilidade térmica da blenda polimérica de PVA e gelatina. Os filmes de gelatina (FG) foram mais rígidos (maior Módulo de Young), enquanto o PVA contribuiu para aumentar a resistência à tração e a capacidade de alongamento da blenda polimérica com a gelatina. O isolamento da antocianina promoveu um efeito plastificante, no qual o FA foi o mais elástico. Quanto a topografia, FB, FF e FA exibiram uma superfície mais heterogênea que o filme de PVA (FPVA) e gelatina controle, devido a incorporação dos extratos de mirtilo que reduziram a interação entre PVA e gelatina. A incorporação de EB e EFA dificultou a interação entre PVA e gelatina diminuindo o teor de umidade do FB e do FA. A solubilidade da blenda

Quando aplicados em embalagens inteligentes os indicadores de FA mudaram de verde para marrom, e o FB de roxo para verde ou marrom, em resposta à degradação de filés de camarão e do peixe, indicando deterioração do camarão e do filé de Tilápia, respectivamente, após 7 dias de armazenamento. Para o camarão, os indicadores FA e FB mudaram de cor concomitantemente ao aumento do pH (7.93) do alimento, sendo esse valor considerado impróprio para consumo segundo a Instrução Normativa nº 23/19, enquanto para o filé de peixe, os valores de pH (6.83) estavam acima dos permitidos pela Portaria nº 1255/62 da RISPOA. O indicador inteligente e sustentável FB foi mais estável durante o armazenamento, sensível e irreversível à mudança de cor, além de mais simples e rápido de produzir, por não requer a etapa de purificação. Dessa forma, recomenda-se o FB como indicador colorimétrico para monitoramento e controle da qualidade de pescados.

Palavras-chave: extrato natural ; mirtilo; filme indicador colorimétrico; poli (vinil álcool); gelatina; embalagem inteligente

LISTA DE SIGLAS E ABREVIATURAS

Sigla/Abreviatura	Significado
AIC	Akaike information criterion
EFA	Anthocyanin extract
EM	Blueberry extracts
EB	Crude extract
DTG	Derivate thermogravimetric
LOD	Detection limit
EAB	Elongation at break
FA	Film with anthocyanin extract
FB	Film with crude extract
FF	Film with phenolic extract
FG	Gelatin film
ΔE	Global color difference
G	Grammage
O	Light absence and oxygen presence
A	Light and oxygen absence
LO	Light and oxygen presence
L	Light presence and oxygen absence
MAPA	Ministério da Agricultura e Pecuária
U	Moisture
EF	Phenolic extract
PVA	Poly (vinyl alcohol)
FPVA	Poly (vinyl alcohol) film
FC	Polymer blend of poly (vinyl alcohol) and gelatin
RISPOA	Regulamento de Inspeção Industrial e Sanitária de Produtos de Origem Animal
S	Solubility
I	Swelling index
TB	Tensile strength at break
TG	Thermogravimetric
TK	Thickness
TVB-N	Total volatile nitrogen base
UTS	Ultimate tensile strength
YM	Young's modulus

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1. INTRODUTION

The growing demand for sustainable and innovative alternatives in the food industry has driven research in biodegradable and intelligent packaging. One strategy proposed is the development of a colorimetric indicator film made of PVA (polyvinyl alcohol), gelatin, and blueberry extract.

The use of polymers such as PVA, a non-toxic, water-soluble, and biodegradable hydrolyzed derivative of polyvinyl acetate (QIAO et al., 2024; YANG et al., 2022), and natural biopolymers such as gelatin, a collagen-derived protein with high oxygen barrier properties and excellent biodegradable film-forming capacity, has shown great potential in food applications (SAI-UT et al., 2021). The combination of PVA with gelatin brings benefits to the packaging industry and the environment by developing polymer blends with good mechanical, thermal, and physicochemical properties, offering an alternative to conventional plastics (OYEOKA et al., 2021). Different methods and different polymer concentrations can be used to form a polymer matrix of PVA and gelatin, as observed in the study by Rashid et al. (2023), Zeng et al. (2019), Oyeoka et al. (2021) and Gomes et al. (2024), thus being able to obtain PVA and gelatin films with different characteristics.

Smart packaging refers to the material that can be used in food to monitor freshness, display quality information, and enhance product and consumer safety while aligning with Industry 4.0 (SCHAEFER; CHEUNG, 2018). In the fishing industry, preserving the sensory and microbiological quality of seafood is essential to ensure food safety and extend shelf life, as these are highly perishable foods that are sensitive to improper processing, transportation, and storage conditions. In this regard, developing intelligent packaging that can monitor seafood quality from the moment of catch to the final consumer is promising and necessary (ZHAI et al., 2017).

Color indicator films are fundamental for advancing the field of intelligent packaging and can provide a solution for seafood quality control. Although efficient indicators developed with chemical dyes exist, they often present limitations in food applications due to potential toxicity (DRAGO et al., 2020; MA; DU; WANG, 2017). Thus, natural plant-based extracts, such as blueberry extract, have been incorporated into polymeric films to add bioactive properties such as antioxidant and antimicrobial effects and to develop color and pH indicators (GOMES et al., 2024; LI et al., 2023).

Blueberry (*Vaccinium spp*.) is widely recognized for its health benefits due to its high anthocyanin content, which are phenolic compounds that alter their chemical structure and consequently change color with pH variations (CAROLINE et al., 2020; KIM et al., 2021).

These pigments appear red under acidic conditions, neutral in color at neutral pH, and blue or yellow in alkaline environments. Ten kinds of anthocyanins carrying malvidin, delphinidin, petunidin, cyanidin four common aglycones from blueberry extract were identified for Liu et al. (2018) and in total, forty-three anthocyanins have been identified for Neuenfeldt et al. (2020), being found cyaninidins, delphinidins, malvidins, petunidins, and peonidins with different ligands. Therefore, blueberry extract emerges as a promising candidate for incorporation into PVA and gelatin films to enhance food safety by monitoring the real-time quality of packaged food, indicating signs of deterioration or contamination, and reducing waste of fresh, consumable food.

This study aimed to investigate the incorporation of three blueberry extracts - the crude extract (EB) and the extracts obtained after purification of the raw extract, phenolic extract (EF) and the anthocyanin extract (EFA) - into PVA and gelatin-based films, focusing on the development of intelligent color-indicating films for seafood monitoring. Specifically, it sought to evaluate the *in vitro* colorimetric behavior (acidic and basic volatile vapors) of the blueberry extracts and the PVA, gelatin, and blueberry extract films: FB (PVA and gelatin film with crude extract), FA (PVA and gelatin film with anthocyanin extract), and FF (PVA and gelatin film with phenolic extract). The study assessed the color sensitivity and reversibility of the films and their stability over time in the presence and absence of oxygen at room temperature. Additionally, it was analyzed the optical, mechanical, physical, chemical, thermal, and topological properties of the films. Furthermore, the present study explored the capacity and effectiveness of FB and FA in monitoring the quality of fresh tilapia fillets and grey shrimp stored under refrigeration (6.5°C) for seven days.

The thesis is structured as follows: Chapter 1 presents a literature review on intelligent packaging, biodegradable polymers, and blueberry extract. Chapter 2 includes an article published in the journal *Food Control* (https://doi.org/10.1016/j.foodcont.2024.110648), in which blueberry extracts and PVA-gelatin polymer blends were developed, and their behavior in the presence of two vapors (ammonia vapor and acetic acid vapor) was studied, along with the characterization of the extracts and films. This chapter was formatted according to the Food Control guidelines. Chapter 3 contains an article, presenting the results of the morphological, chemical, physical, thermal, and mechanical properties of the films. Finally, Chapter 4 features an article, evaluating the stability over time, as well as the presence and absence of light and oxygen, the reversibility and colorimetric sensitivity, and the effectiveness of the color indicators FB and FA when applied to fresh tilapia fillets and grey shrimp.

Despite various studies exploring the use of biopolymers as an alternative to conventional plastics and the use of natural plant extracts instead of synthetic chemical compounds (ETXABIDE; KILMARTIN; MATÉ, 2021; FREITAS et al., 2020; SGANZERLA et al., 2021; TEIXEIRA et al., 2022a; WANG et al., 2023), the application of three different fractions of blueberry extract has not yet been explored, particularly for the development of colorimetric indicators for food packaging. Moreover, the effects of natural extract additions on film properties and their application for seafood monitoring need further understanding, justifying the relevance of this research. Therefore, the use of blueberry extracts, rich in antioxidant and antimicrobial compounds, can provide an innovative approach for real-time food quality monitoring by detecting signs of deterioration or contamination, using PVA and gelatin as polymer matrices for film formation reflects the pursuit of biodegradable and food-safe materials.

2. BIBLIOGRAPHICAL REVIEW: The potential of blueberry extracts in smart packaging biodegradable polymer-based: A Review

1. Introducion

A large amount of plastic is used and discarded every day, generating a tons of waste and several environmental problems, with a focus on plastic food packaging (JESUS, 2021; KALPANA et al., 2019; MA et al., 2018).

Food packaging is an essential component that serves as a barrier and protection against contamination, changes caused by the external environment, adulteration, and mechanical damage, ensuring the quality, integrity, and safety of food products (KALPANA et al., 2019). Furthermore, the functions of food packaging include containment, physical protection, convenience, product identification, and communication between producer and consumer (JESUS, 2021; KALPANA et al., 2019; MA et al., 2018; ZENG et al., 2019a). It is a preparation system for transportation, distribution, storage, sale, and subsequent final use (JESUS, 2021; KALPANA et al., 2019; MA et al., 2018).

Food packaging materials are typically made of synthetic polymer matrices based on petroleum, which dominate the market due to their low cost and simple accessibility. These matrices are composed of polyolefins (polyethylene and polypropylene, polyvinyl chloride, polystyrene, polyethylene terephthalate and ethylene vinyl alcohol (EVOH), which are responsible for providing a significant barrier to water. However, they present some limitations with petroleum resources and the not biodegradation, which increase ecological and cost-benefit concerns (LUZI et al., 2019). Therefore, the development of environmentally friendly short-lifespan packaging is a challenge that aims to minimize plastic use, thereby reducing environmental impacts, without compromise food quality ant shelf life (LUZI et al., 2019).

Food waste is also one of the main issues for international organizations, besides the ethical and economic concern, the worry about the depletion of limited natural resources (POYATOS-RACIONERO et al., 2018). Percentages of food loss are highly relevant in products such as cereals, dairy, roots, fish, meat and tubers, with more than 300 million tons per year (FAO, 2022). Therefore, reducing waste from households and businesses is one of the concerns of the food sector and the population (POYATOS-RACIONERO et al., 2018). According to the Food and Agriculture Organization (FAO), milk waste at the consumption level represents approximately 40-65% and for fishery products 35% of total food waste in industrialized regions (Europe, North America, Oceania, and industrialized Asia).

The basic functions of food packaging such as containment, physical protection, convenience and communication are not sufficient for fresh food. These fresh food materials with high protein and fat content, as fish and meat, are perishable and require greater assurance and real-time freshness monitoring. With emphasis on the fish that degrades faster than meat (POYATOS-RACIONERO et al., 2018). Consequently, Among the appropriate strategies to address this challenge, smart packaging is an interesting tool.

The growing interest of the global population in the state of the environment, especially concerning the production of toxic waste, pollution, contamination, depletion of natural resources, and environmental degradation, drives the scientific community towards the development of packaging that meets the ideals of the circular economy, aiming to reduce waste and residues, and provide more sustainable products and processes.

The application of plant extracts rich in anthocyanins from different sources as smart agents in different biopolymers (such as starch, chitosan, carboxymethyl-cellulose, polyvinyl alcohol, gelatin, and agar, among others) for the development of smart films for food (especially fish, seafood, pork, and milk) is an effective alternative to monitor the quality and safety of these foods (OLIVEIRA FILHO et al., 2021).

2. Smart Packaging

Smart packaging has been extensively researched with the aim of reducing the amount of food waste, which is one of the main issues faced by international organizations, not only because it is an ethical and economic concern but also because it leads to the depletion of limited natural resources (POYATOS-RACIONERO et al., 2018).

In addition to the basic function of food protection, smart packaging monitors and shares information in a convenient, fast, dynamic, and non-destructive way to consumers about the actual quality of food, such as meat, seafood, dairy products, as well as fruits and vegetables, indicating the freshness and stage of deterioration of the food without the need to open the packaging (KALPANA et al., 2019; KOSHY et al., 2021; LE et al., 2019; TEIXEIRA; SOARES; STRINGHETA, 2021).

Consumers are increasingly aware of safety and opt for high-quality and convenient foods due to the fast-paced lifestyle, besides the concerns about the environment (TEIXEIRA; SOARES; STRINGHETA, 2021).

With the use of smart packaging, food can be discarded when it is no longer suitable for consumption, simply based on the visual appearance of the packaging, in addition to providing alerts to consumers about how much time remains for consumption. In this way, these packages

reduce waste and improper food consumption, serving as a tool for selecting safe food for the consumer in retail (KALPANA et al., 2019; KOSHY et al., 2021).

Usually, an expiration date is printed on food products, which are subject to various physical, chemical, and microbiological actions. On the other hand, smart packaging provides the actual shelf life of the food, rather than a validity based on numerical and statistical parameters, as the product may have been exposed to various adverse and unfavorable conditions that may not have been considered during the shelf-life study (PASCHOA, 2016).

One of the limitations of smart packaging by industries is the high the cost, challenging the analysis of their cost-benefit (KALPANA et al., 2019). There is also a concern regarding food safety when smart packaging materials come into direct contact with food (KALPANA et al., 2019). This raises the issue of particle migration into the food material (KALPANA et al., 2019). Additionally, these packaging materials may have limitations regarding their mechanical, barrier, and thermal properties, which challenge their commercial use (JESUS, 2021).

Thus, to approve and assess the effectiveness of a smart packaging, it must be considered that food safety cannot be compromised (POYATOS-RACIONERO et al., 2018). The systems must be robust and avoid any false negatives (samples that appear safe but are actually unsuitable) and, in addition, they must have the lowest possible false positive rate (samples that seem unsafe but are actually safe for consumption), in order to maximize their efficiency in relation to food waste (POYATOS-RACIONERO et al., 2018). Validation studies for packaging should include large sample sizes and be tested under as many conditions as possible (POYATOS-RACIONERO et al., 2018), and the need for research into smart tools for liquid foods is emphasized (KALPANA et al., 2019).

The devices used in smart packaging, such as indicators, sensors, or data carriers, are inserted or incorporated into the body of the packaging in the form of labels and tags, so they can interact with the internal and external components of the food and the environment in which it is conditioned, providing as a result an immediate response that correlates with the physical, chemical, and biological properties of the food (MA et al., 2018; POYATOS-RACIONERO et al., 2018; YAM, 2012). This information is qualitative and is usually presented as an immediate visual change, with most indicators relying on visible color changes, classified as colorimetric indicators (MA et al., 2017; O'GRADY; KERRY, 2008).

3. Colorimetric indicator films

Colorimetric indicator films can manufacture smart packaging that allows communication with the consumer (EZATI; RHIM, 2020c; KOSHY et al., 2021; ZENG et al., 2019a) through visible color changes (ZENG et al., 2019a). Indicator films consist of a solid support matrix that immobilizes a dye sensitive to changes in the environment, such as pH (LUO et al., 2023; ZENG et al., 2019a).

In general, pH is considered the most important factor directly related to food quality (LUO et al., 2023) that can change through the presence of degradation compounds. Fish, shellfish, meat, and milk contain large amounts of proteins and fats, which produce volatile or non-volatile organic acids and nitrogenous substances after microbial or enzymatic decomposition, causing significant changes in the pH of fresh foods (LUO et al., 2023). Therefore, several studies have been conducted to develop pH-sensitive colorimetric indicators. A pH-responsive colorimetric indicator effectively, simply, and quickly monitors real-time changes in the quality (freshness or spoilage) of perishable food products, as well as inspecting their shelf life, displaying various colors at different pH levels, whether acidic or basic, allowing consumers to reject consumption (MA et al., 2018; TEIXEIRA; SOARES; STRINGHETA, 2021).

Various synthetic dyes are used for the production of quality indicators, as they are highly sensitive to pH changes and easily produce distinguishable color changes (TEIXEIRA et al., 2022a). However, synthetic dyes have potential toxicity and pathogenicity, posing a significant threat to consumer health, as they can migrate into food (CAO et al., 2019). Thus, the use of synthetic dyes in food colorimetric indicator films should be avoided to ensure safety (ZHANG; LU; CHEN, 2014).

Therefore, researchers have currently been using natural dyes, extracted from fruits and vegetables, to develop colorimetric indicator films due to their non-toxic, natural, and eco-friendly characteristics, with an emphasis on anthocyanins (EZATI; RHIM, 2020b; LUO et al., 2023; PRIETTO et al., 2017).

Natural pigments can provide immediate qualitative information through visual color changes caused by the structural alteration of the pigment in response to environmental changes, such as pH alteration that occurs with food spoilage (AYDOGDU EMIR, 2023; JAYAKUMAR et al., 2019; KALPANA et al., 2019). Furthermore, due to the natural antioxidant properties of anthocyanins, smart packaging can act as active packaging, offering dual functionality (SGANZERLA et al., 2021).

Sganzerla et al. (2021) produced and characterized a biodegradable film based on carboxymethylcellulose and anthocyanin-rich extract from blackberry (*Morus nigra* L.), and it was concluded that the film changed color according to the pH of the exposed medium.

Agunos, Mendoza and Rivera (2020) developed a colorimetric food spoilage indicator film by incorporating anthocyanin extracted from mangosteen peel into a chitosan/PVA matrix, sensitive to ammonia vapor, which caused a color change from reddish pink to light yellow due to the increase in pH.

Moreover, anthocyanins from *Clitoria ternatea* were incorporated into a polymeric starch base with carbon nanotubes to monitor the spoilage of pork meat (KOSHY et al., 2021). Also, pH-responsive indicator films based on methylcellulose/chitosan nanofibers and barberry anthocyanins were developed for real-time monitoring of meat freshness through color change (ALIZADEH-SANI et al., 2021). Besides, pH-sensitive anthocyanins found in purple potatoes or roselle were incorporated into chitosan and polyvinyl alcohol matrices with zinc oxide nanoparticles to produce smart films (LIU et al., 2021)

Ma et al. (2018) prepared a visually responsive smart film using PVA, chitosan nanoparticles (CHNPs), and blackberry extracts. The CHNPs were prepared to improve the mechanical properties of the films. The film was tested for monitoring fish. The film's color changed from red to green due to fish degradation (increase in pH), and it could be used as a intelligente packaging label to detect food unfit for consumption.

In addition, a pH-sensitive colorimetric indicator film with barberry anthocyanins and a matrix of methylcellulose and chitosan nanofibers was developed by Alizadeh-sani et al. (2021). The film showed an apparent color change, from reddish pink to pale peach and yellow in response to pH changes, and from pink to pale green and yellow in the presence of ammonia gas, making it suitable for indicating pH changes in foods, the formation of volatile nitrogen compounds, and food spoilage. Furthermore, the indicator film was successfully used to monitor and inform consumers in real time about the quality and safety of lamb meat and possibly other protein-rich foods.

Weston et al. (2020) extracted anthocyanins from purple cabbage, and they exhibited a colorimetric response from blue to red when exposed to lactic acid, indicating microbial spoilage in milk. The extract was incorporated into an agarose matrix to discriminate the milk quality, distinguishing between fresh and spoiled milk. Future work incorporating this colorimetric indicator into milk packaging has the potential to reduce food waste, assisting consumers and food management at retail.

In the study by Zeng et al. (2019a), anthocyanin extracts from mulberry residues were incorporated into a gelatin/polyvinyl alcohol matrix to prepare a colorimetric indicator film. The interaction between the anthocyanin extracts and the gelatin/polyvinyl alcohol matrix was dominated by hydrogen bonding and electrostatic interaction, which enhances the stability of the anthocyanin. The addition of anthocyanin extracts improved the mobility of the film matrix, resulting in decreased tensile strength and increased elongation at break of the films. The anthocyanins in the film provide pH-sensitive responses and are reliable in monitoring volatile nitrogenous compounds formed during the fish spoilage process. Therefore, this colorimetric indicator film holds great potential as an effective, simple, sustainable, biodegradable, and non-toxic method for monitoring food spoilage.

Several advanced methods, such as electrospinning, casting, and immobilization with nanoparticles, have been used to manufacture pH-sensitive colorimetric indicator films for use in intelligent packaging. These have been widely used to monitor the freshness or deterioration of fruits, vegetables, fish, meat, milk, and seafood (LUO et al., 2023).

Recently, several polymeric and biopolymeric materials have been used as solid matrices for colorimetric indicators, with a focus on naturally occurring polymers such as starch, cellulose, chitosan, chitin, gelatin, and κ-carrageenan (AYDOGDU EMIR, 2023; LUO et al., 2023). Therefore, there are many options for developing a biodegradable indicator since the sources of natural indicators and the bio-based availability are vast, and their combination needs to be investigated to understand the color transition and the impact in polymeric properties.

4. Biodegradable Polymers

Polymers are macromolecules composed of many (tens of thousands) repeating units, known as mers, linked by covalent bonds, with each monomer being able to bond with at least two other monomers for the polymerization reaction to occur (JESUS, 2021). Most packaging materials are made from polymers derived from petrochemical sources due to their high availability at relatively low cost, and mechanical properties that allow for easy handling with controlled heating. However, they are derived from non-renewable sources and are not biodegradable, creating environmental problems due to the difficulty in waste management and recycling (JESUS, 2021; MA et al., 2018).

Biopolymers are considered 'green' polymer matrices made from natural raw materials through synthetic routes (LUZI et al., 2019). The use of biopolymers is a sustainable development approach that aids in environmental preservation and has gathered considerable

attention in the last two decades (OYEOKA et al. 2021). Biopolymers or biodegradable polymers from renewable sources, such as polysaccharides and proteins, represent only 5 to 10% of the current market, but they have been favored by researchers due to their biodegradability and feasibility and they are being studied and considered as an alternative to reduce the excessive use of plastics and, consequently, to mitigate environmental problems (JESUS, 2021; KOSHY et al., 2021; LUZI et al., 2019; MA et al., 2018). Biodegradable polymers are polymers which can be degraded by enzymatic action of living organisms, such as bacteria, yeasts and fungi (OYEOKA et al. 2021).

However, the properties of films prepared from natural sources must be improved to compete with petroleum-based polymers, especially mechanical and barrier properties, and those related to water affinity (LUZI et al., 2019). Moreover, molecular weight, physical properties (crystallization phenomenon and degree of crystallization), viscoelasticity, and rheological characteristics may present other disadvantages (LUZI et al., 2019).

To overcome these limitations, it is necessary to make modifications or adjustments during the processing stages to modulate and improve the physical and functional properties of these films, such as chemical or physical changes (crosslinking). Biodegradable polymers can be blended to combine characteristics and offer functionality and/or be added with fillers and components (compatibilizers or plasticizers) (LUZI et al., 2019; MA et al., 2018).

Plasticizers are low molecular weight agents incorporated into films to increase flexibility and processability. They cause an increase in the free volume of the polymer structure or the molecular mobility of the matrix, a decrease in the proportion of crystalline regions to the amorphous region, and a reduction in the glass transition temperature (JESUS, 2021). To form some biodegradable films, such as gelatin and PVA film, the polymer is mixed with a solvent and a plasticizer to improve the malleability of the film, heated to exit the crystalline state and then slowly dried so that the chains organize and solidify (JESUS, 2021; GOMES et al., 2024).

The main cost drivers for the production of these biodegradable materials include the cost related to mobilizing biomass waste, technical and scientific innovations, and the lack of an ecologically correct biological base creation chain (LUZI et al., 2019). Moreover, biodegradable polymer films require careful post-consumption management and efficient design to enable biodegradation (NARANCIC et al., 2018).

Biodegradable films developed with natural macromolecules of biological origin can be considered one of the promising alternatives to reduce and/or limit the environmental impact in relation to petrochemical polymer matrices (JESUS, 2021; KOSHY et al., 2021; PEELMAN et al., 2013; SGANZERLA et al., 2021). These macromolecules are obtained by processing

renewable resources (vegetal and animal waste) and offer several positive aspects, such as environmental benefits, disintegrable, degradability, potential for recycling polymer waste, absence of toxic components, and high biocompatibility (LUZI et al., 2019).

Gelatin is a thermally fusible macromolecule made from the partial or total hydrolysis of collagen and applied in various fields due to its low cost (DJAGNY; WANG; XU, 2001). It contains a large amount of proline and hydroxyproline residues, which have an affinity for polyphenols (GARCÍA-ESTÉVEZ et al., 2017).

Gelatin is a colorless, edible, renewable, biodegradable, non-toxic, and water-soluble biopolymer (DJAGNY; WANG; XU, 2001; LUO et al., 2022). Due to its availability, film-forming ability, biocompatibility, flexibility, good mechanical properties (with some limitations), and gas barrier properties, gelatin is a biopolymer that is widely studied in the production of biodegradable films (LUO et al., 2022; OYEOKA et al., 2021). However, gelatin-based films can be excessively rigid and brittle, sensitive to humidity, and easily dissolve, swell, or crack when in contact with water, creating limitations for their use in food packaging (LUO et al., 2022; TYMCZEWSKA et al., 2022).

In light of this, polyvinyl alcohol (PVA) has been widely used in combination with natural proteins to improve the mechanical properties of biodegradable polymer films (MA et al., 2018; MA; DU; WANG, 2017; ZENG et al., 2019a). Polyvinyl alcohol (PVA) is a synthetic polymer that is transparent, derived from non-renewable sources, non-toxic, with high biocompatibility, biodegradability, and promising for film production (MALI et al., 2019; OYEOKA et al., 2021). It is a water-soluble polymer bonded only by carbon-carbon links and has a high density of hydroxyl groups on its side chains (ZHAO et al., 2022). On an industrial scale, PVA is obtained by complete or partial hydrolysis of polyvinyl acetate, in which the acetate groups are removed. It is classified as highly hydrolyzed (98-99%) and partially hydrolyzed (above 88%) (TANG; ALAVI, 2011).

Polymeric blends formed by PVA and gelatin are macroscopically homogeneous, colorless, and without opacity (KIM; ROY; RHIM, 2022; MARIA et al., 2008) and have shown potential as biodegradable packaging materials (HAGHIGHI et al., 2021), as mixing these polymers results in a more stable and resistant food packaging (TYMCZEWSKA et al., 2022).

Polymeric blends of gelatin and PVA, combined with anthocyanin extracts, change their color under the presence of acidic and basic volatile compounds. These blends can be used as colorimetric indicator films to monitor the freshness of protein-rich foods during storage (GOMES et al., 2024; KAMER et al., 2022; ZHANG et al., 2019).

5. Blueberry Extract

Blueberry (*Vaccinium myrtillus*) is a fruit native to the Northern Hemisphere, with the United States being the largest global producer and consumer of this fruit (CARPENEDO; RASEIRA; FRANZON, 2022; ROCHA et al., 2018b). In Brazil, the first records of the production of this species were in 1983, in Pelotas (RS), with the country being a small producer (CARPENEDO; RASEIRA; FRANZON, 2022). The fruit is gaining more and more space in the Brazilian market, as it is now better known, accessible, and available.

The prospects for blueberry cultivation in Brazil are very promising, proving to be viable in various regions, such as the city of Barbacena in Minas Gerais, showing an increasing scale of planting and consumption, as well as genetic improvements to ensure better adaptation of cultivars to Brazilian regions, with good productivity and fruit quality (CARPENEDO; RASEIRA; FRANZON, 2022).

Blueberries have a high concentration of phenolic compounds, such as anthocyanins, flavonoids, and phenolic acids, making them a source of bioactive compounds with high antioxidant capacity and potential health benefits (NEUENFELDT et al., 2022; ROCHA et al., 2018b; RODRIGUES et al., 2011). The levels of phenolic compounds vary with the blueberry cultivar, production location (ROCHA et al., 2018b; RODRIGUES et al., 2011), and degree of ripeness (BERNAL-GALLARDO et al., 2022).

The extraction of bioactive compounds from plant tissues can be performed through ultrasound-assisted extraction, which stands out for its high energy efficiency, shorter time, and higher yield of extracted bioactive components (BARRETO et al., 2020; DOS SANTOS et al., 2021; HU et al., 2019; ROCHA et al., 2018a). Ultrasound-assisted extraction is a green method based on the polarity and solubility of active components. The production of high-frequency ultrasonic mechanical waves results in cavitation, a phenomenon responsible for the formation and collapse of bubbles, which generate high-pressure and temperature points. This leads to significant rupture of cell structures and solvent penetration, resulting in high mass transfer rates from the plant tissue to the extraction solution. This process allows the easy extraction of a bioactive-rich extract (BAI et al., 2023; BARRETO et al., 2020; BENDICHO et al., 2012; DE FREITAS et al., 2015).

The solvents used in the extraction of anthocyanins are not specific, and therefore, other compounds may also be extracted, concentrated, and measured. As a result, the qualitative analysis and quantification of anthocyanins in raw extracts can be delicate (RODRIGUEZ-SAONA; WROLSTAD, 2001a). The crude extract has low purity and stability (BAI et al., 2023), making the purification of raw extracts often necessary to remove interfering non-

phenolic compounds. This process also allows for the separation of these extracts (RODRIGUEZ-SAONA; WROLSTAD, 2001a), thereby obtaining purified phenolic extracts in two fractions: the anthocyanin phenolic extracts and the extracts containing other compounds (COKLAR; AKBULUT, 2017).

Common methods for separating and purifying anthocyanins include resin purification, chromatography, liquid-phase extraction, and solid-phase extraction (BAI et al., 2023). Purification using solid-phase extraction allows for the removal of various interferents and high efficiency in fractionating the extracts (RODRIGUEZ-SAONA; WROLSTAD, 2001a). Mini-columns containing C18 chains bonded to silica retain hydrophobic organic compounds (such as anthocyanins and phenolics) while allowing matrix interferents, such as sugars and acids, to be removed with acidified water (RODRIGUEZ-SAONA; WROLSTAD, 2001a). Washing the retained pigments with ethyl acetate removes polyphenolic compounds (RODRIGUEZ-SAONA; WROLSTAD, 2001a). Subsequently, the relatively pure anthocyanin extract can be eluted from the column with lightly acidified methanol (RODRIGUEZ-SAONA; WROLSTAD, 2001a).

Natural extracts obtained from fruits, such as blueberries, are rich in anthocyanins, a natural dye capable of responding to pH changes through transformations that occur in their structures, which consequently alters the wavelength of light reflected, changing their color (GOMES et al., 2024; JAYAKUMAR et al., 2019; KOSHY et al., 2021). Anthocyanins are found in various fruits, flowers, and vegetables, with a broad range of colors including red, purple, and blue. They are water-soluble, non-toxic, and have high antioxidant capacity, making them a promising substitute for synthetic food antioxidants and dyes (MA et al., 2018; WANG; JUNG; ZHAO, 2017).

The main anthocyanins found in blueberries are cyanidin-3-O-glucoside, delphinidin-3-O-galactoside, delphinidin-3-O-glucoside, delphinidin-3-O-arabinoside, cyanidin-3-O-galactoside, cyanidin-3-O-arabinoside, petunidin-3-O-galactoside, petunidin-3-O-glucoside, petunidin-3-O-arabinoside, peonidin-3-O-galactoside, peonidin-3-O-glucoside, peonidin-3-O-arabinoside, malvidin-3-O-galactoside, malvidin-3-O-glucoside, malvidin-3-O-arabinoside, and malvidin-3-O-xyloside (JARA et al., 2019; PAES et al., 2014; PIRES et al., 2020).

In addition to anthocyanins, blueberries also contain other phenolic compounds such as the flavonoids pterostilbene, epicatechin, catechin, myricetin, quercetin, procyanidins, and phenolic acids like syringic acid and chlorogenic acid (BAI et al., 2023) and acid 5-p-coumaroylquinic (PIRES et al., 2020) which may be sensitive to pH changes.

Thus, natural extracts are being widely used as natural colorant for colorimetric smart indicators to monitor food quality (EZATI; RHIM, 2020b; KOSHY et al., 2021; TEIXEIRA et al., 2022; ZHAO et al., 2022). The use of smart packaging with anthocyanins incorporated into polymer matrices provides an innovative tool for obtaining visual and real time information about the condition of the food (MA et al., 2017; YONG et al., 2019b). Additionally, these natural antioxidants can be released from the packaging into the food matrix, reducing degradation reactions and acting as an active packaging (SGANZERLA et al., 2021; YONG et al., 2019a).

6. Final considerations and future perspectives

The future of food packaging lies in packages that can do more than just protect and contain the products; it is moving towards biodegradable and sustainable packaging that reduces food waste and environmental pollution, besides providing food safety for consumers. In this sense, smart packaging made from biodegradable polymer bases and enhanced with natural extracts is becoming increasingly valued, studied, and explored.

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3. ARTICLE 1: Gelatin/polyvinyl alcohol films incorporated with different blueberry extracts as potential colorimetric indicators to detect acidic and basic vapors

Abstract

Blueberry extract is rich in phenolic compounds and anthocyanins, which can undergo color changes when subjected to different pH conditions. This feature makes it liable to be incorporated into polymeric matrices to form colorimetric indicator films. In this sense, films based on gelatin and polyvinyl alcohol (PVA) were elaborated with raw blueberry extracts (EB), purified fractions of phenolic extract (EF) or anthocyanin extract (EFA). Their color sensitivity to acidic and basic vapors was investigated. Higher levels of phenolic compounds and total anthocyanins were observed for EB and EFA. The EFA was more sensitive to acidic and basic vapors, with an intense visual colorimetric change attributed to the anthocyanins. The higher absorbance in the 400 nm wavelength region observed in both EF and EB spectra was possibly due to the presence of yellow phenolic compounds. Corroborating to the findings for the extract, the EFA film presented a tone shift over time and a higher overall color difference, completely changing from a greenish color to purple under acidic vapors. However, the EB film has shown to be more susceptible to basic conditions, exhibiting differences in h° values over time under basic vapors. Therefore, PVA/gelatin films incorporated with EB are recommended for the colorimetric indication of nitrogenous bases, and the EFA-added films for volatile acids. Based on the results, films incorporated with EB or EFA can be used to monitor, detect, and report the presence of acidic and basic vapors from protein food degradation.

Keywords: Purified blueberry extract; Blueberry crude extract; Colorimetric change; Intelligent packaging; Sustainability.

1. Introduction

Blueberries (*Vaccinium myrtillus*) are native to the northern hemisphere, with the United States being its largest producer and consumer worldwide. These fruits present a high concentration of phenolic compounds, such as anthocyanins, flavonoids, and phenolic acids, being considered a source of bioactive compounds with high antioxidant capacity (Neuenfeldt et al., 2022). However, to harness their full potential as a source of phenolic compounds on an industrial scale, it is imperative to enhance the fruit yield, optimize harvest efficiency, and explore innovative extraction techniques (Bai et al., 2023).

During extraction of blueberries' bioactive compounds, due to the use of nonspecific solvents, a wide range of substances are obtained (e.g., anthocyanins, phenolic compounds, flavonoids, and phenolic acids) and then concentrated to form the crude extract (Rodriguez-Saona & Wrolstad, 2001). The crude extract has low purity and low stability (Bai et al., 2023), making its purification often necessary to remove non-phenolic compounds. Purification also allows the separation of bioactive compounds from these extracts (Rodriguez-Saona & Wrolstad, 2001), thus obtaining two fractions: the phenolic anthocyanin extract and a second extract comprehending the other compounds, including phenolic acids, flavonols, and flavan-3-ols (Coklar & Akbulut, 2017; Pires et al., 2020).

The phenolic compounds and anthocyanins found in a variety of vegetable species, such as red cabbage, açaí, and blueberries, can change color in response to varying pH conditions (Freitas et al., 2020; Silva et al., 2022; Teixeira, de Oliveira, et al., 2022). As a result, they can be incorporated into polymeric matrices and manufacture novel indicator films (Aydogdu Emir, 2023), contributing to the advancements and innovations in materials development.

Indicator films serve as systems capable of informing the consumer about the inherent variations through colorimetric changes (Teixeira et al., 2021). Particularly in the context of packaged food, integrating these films into packaging systems enables the communication of important information regarding the food's condition to the consumers, including ripeness, gas or volatile compounds presence, temperature abuse, and overall food quality. To develop indicator films, the polymer matrix is used as a base structure for the colorimetric compounds, such as blueberry phenolic extracts (Kim et al., 2022).

Following the sustainability trend, indicator films can also be produced using biopolymers (starch, cellulose, chitosan, alginate, dairy proteins, soy and gelatin) and biodegradable synthetic polymers (polyvinyl alcohol - PVA, polycaprolactone - PCL), as an alternative to traditional non-biodegradable packaging materials (Hernández-García et al.,

2021; Sai-Ut et al., 2021). Polymeric blends based on PVA and gelatin are macroscopically homogeneous, colorless and without opacity (Kim et al., 2022). Gelatin and PVA films have been extensively studied as potential biodegradable packaging materials (Haghighi et al., 2021) since their blending enables the obtention of a more resistant material (Tymczewska et al., 2022).

Polymeric blends of gelatin and PVA enhanced with anthocyanin extracts can alter their color in response to the production of acidic and basic volatile compounds. This property allows them to be used as colorimetric indicator films for monitoring the freshness of protein-rich foods during storage, as the degradation of these kind of products releases volatile compounds that could be effectively detected (Kamer et al., 2022; Zhang et al., 2019).

To the present date, there is no research published in the literature database that compares the chromatic transition effect between crude blueberry extract (EB) and the extracts obtained from the purified fractions: anthocyanin extract (EFA) and phenolic extract (EF). Moreover, there are no reports on the incorporation of these extracts into polymeric blends of PVA and gelatin. Hence, this work aimed to fill this gap and develop PVA/gelatin films incorporated with raw blueberry extract or purified fractions. Their impact on colorimetric sensitivity under acidic and basic conditions was further investigated, simulating the detection of volatiles released during food spoilage.

2. Materials and Methods

2.1 Materials

Blueberry (*Vaccinium myrtillus*) Probst fruits were obtained from the fruit-producing company Quali Fresh (Barbacena, Minas Gerais, Brazil) and stored at -18 ± 2 ºC until analysis. The polymers used in the film preparation were PVA (Sigma Aldrich Co., St. Louis, MO, USA), degree of hydrolysis of +99% and molecular weight (Mw) of 85,000 to 124,000, and 99% hydrolyzed, colorless, flavorless gelatin from the Dr. Oetker brand (Lot No. L346036A 22N), food grade.

All chemical reagents used in this study were of analytical grade. The following reagents were acquired from Sigma Aldrich Co. (St. Louis, MO, USA): Folin-Ciocalteu Reagent, 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox), ammonia and gallic acid. Glycerol, ethyl acetate, and acetic acid were obtained from Vetec Química Fina Ltda (Recife, PE, Brazil). Ethyl alcohol, methyl alcohol, and sodium chloride were acquired from Merck

(Taufkirchen, Bavaria, Germany). The quantitative paper filter (Unifil, Ø 11) was obtained from Dinâmica (São Paulo, SP, Brazil).

2.2 Methods

2.2.1 Preparation of aqueous concentrated EB

The extraction of phenolic compounds was carried out according to the methodology described by Rocha et al. (2018), with modifications. Briefly, the blueberry was crushed in a mixer (RI1602, Philips Walita, Brazil), mixed with 70% ethanol (v/v), in a 1:10 (w/v) ratio. The mixture was acidified to pH 2.0 with hydrochloric acid (HCl) (37% v/v) and subjected to sonication in an ultrasound bath (Elmasonic TI-H10, Elma, Singen, BW, Germany), at 45 kHz (40 ± 2 °C, 50 min). Subsequently, it was filtered (Whatman No. 1 filter paper) under vacuum and concentrated in a rotary evaporator (RV 10 digital V, IKA, Staufen, BW, Germany), 100 rpm, at 40 ± 2 °C, until the complete ethanol evaporation.

2.2.2 Purification of aqueous concentrated blueberry extract

Purification was done using a C18 separation cartridge (Sep-Pak Vac 35 cc, Waters, Milford, USA), under vacuum. First, the cartridge was conditioned with 50 mL of acidified methanol (0.01% HCl) and 50 mL of acidified distilled water (0.01% HCl). Next, a 50 mL aliquot of the concentrated blueberry extract was loaded into the cartridge. The removal of other compounds (such as sugars) was done with 50 mL of acidified distilled water (0.01% HCl). To obtain the extract rich in phenolic compounds, an additional passage of 50 mL of ethyl acetate was performed, leaving the anthocyanins adsorbed and obtaining the purified phenolic extract (EF). To remove the adsorbed anthocyanins and obtain the purified anthocyanin extract (EFA), 50 mL of methanol was loaded into the cartridge (Noratto et al., 2010; Rodriguez-Saona & Wrolstad, 2001) and the anthocyanins were eluted.

2.2.3 Quantification of phenolic compounds

It was performed according to the Folin-Ciocalteau spectrophotometric method (Singleton & Rossi Jr, 1965). In test tubes, 0.6 mL of each sample was added separately, with subsequent incorporation of 3 mL of Folin-Ciocalteau reagent and 2.4 mL of Na_2CO_3 7.5%. The mixture was stored in the dark for 1 h. Absorbance was measured at 760 nm on a UV-Vis spectrophotometer (UV-M51, Bel Photonics, Monza, Italy). Quantification was performed using a gallic acid standard curve and the results were expressed as mg of gallic acid equivalent (AGE) per 100 g of blueberry.

2.2.4 Quantification of total anthocyanins

It was carried out by the differential pH method, according to Fuleki and Francis (1968). Absorbance was measured in a spectrophotometer, at pH 1.0 and pH 4.5, in wavelengths of 520 nm and 700 nm. The results were expressed as mg of cyanidin-3-glucoside equivalent (cy-3-glu) per 100 mg of blueberry (mg cy-3-glu/100g).

2.2.5 Sensitivity test of extracts in the presence of volatile acids or bases

The absorbance of the extracts was determined using a UV-Vis spectrophotometer, at wavelengths of 400, 420, 440, 460, 480, 500, 520, 540, 560, 580, 600, 620, 640, 660, 680 and 700 nm, before and after exposure to glacial acetic acid or ammonia vapor, for 1 h at 25 ºC. To produce acidic and basic vapor, a beaker containing 500 µL of glacial acetic acid or ammonia solution was deposited inside the desiccator with the extracts (De Oliveira et al., 2015; Ma et al., 2017).

2.2.6 pH

The pH of the blueberry extracts was determined according to the 017/IV method of the Adolfo Lutz Institute (2008). The pH of the crude, purified phenolic, and purified anthocyanin extracts were measured directly before and after exposure to acetic acid and ammonia, using a pHmeter (PG 1800, Gehaka, Brazil), at 25 ± 2 ºC.

2.2.7 Production of colorimetric indicators films

The films were produced by mixing two suspensions: 2% gelatin (suspension A) and 2% PVA (suspension B) (Maria et al., 2008). Suspension A was dispersed in water at 60 ± 2 °C, and suspension B was dispersed in water at 80 ± 2 °C, for 5 h. The suspensions were then mixed and kept under stirring (300 rpm) until cooled to 40 ± 2 °C. Glycerol (30% w/w, based on polymer mass) and extracts (40 mL) were added to the suspension, with the obtention of 3 films: (i) EB film, incorporated with crude blueberry extract, (ii) EF film, incorporated with phenolic blueberry extract, and (iii) EFA film, incorporated with blueberry anthocyanin extract. The control film (without extract addition) was prepared with water (40 mL). The suspensions were homogenized for 20 min, at 25 ± 2 ºC, with magnetic stirring (300 rpm), in order to produce film-forming suspensions. Subsequently, they were placed in an ultrasound bath (40 kHz) for 15 min to remove air bubbles and poured onto glass plates (33 cm x 9) cm² to dry (25 ± 5 °C, RH 62 ± 5%, for 24 h, and 20 ± 2 °C, RH 53 ± 5% for 72 h in a climatic chamber) (420-CLDTS

300, Ethik Technology, Brazil). The films were vacuum packed (200S, Selovac, Brazil) in polyethylene and nylon bags, wrapped in aluminum foil and stored at 20 ± 2 ºC, for subsequent analysis.

2.2.8 Experimental design

The experiment was conducted in a completely randomized design. To evaluate the colorimetric transition of polymer blends under exposure to volatile acids and bases, two factors were studied, namely: the type of blueberry extract, at 3 levels (crude extract 'EB', anthocyanin extract 'EFA', and phenolic extract 'EF'), and the exposure time to acidic and basic vapors, in 8 levels (0, 4, 8, 12, 16, 20, 24, and 28 min). Three replications were performed for each time, and for each PVA and gelatin film (EB, EFA, and EF), totaling 72 experimental units.

2.2.9 Colorimetric sensitivity of films to volatile acids and bases over time

The colorimetric change property of the films when exposed to acid or volatile bases was investigated as proposed by Freitas et al., 2020. The films were cut into squares pieces (2 x 2 cm²), placed in petri dishes, and kept inside desiccators with a controlled RH of 75% (NaCl), for 24 h. After the conditioning time, a beaker containing 500 µL of glacial acetic acid or ammonia solution was placed inside the desiccator with the films.

The color of the films was characterized using a colorimeter (Color Quest XE, Hunter lab, USA), under the following conditions: D65 illuminant, 10º observation angle, and CIELAB system (L*, a*, b*, h°, C*). The colorimetric coordinates (L* luminosity (0 = black and 100 = white), a* intensity from green (-) to red (+), and b* intensity from blue (-) to yellow (+)) were determined every 4 min, for 28 min, to evaluate the colorimetric change profile of the colorimetric indicators. The analysis was carried out for 28 minutes based on preliminary tests, since there were no changes in color under the conditions established after 28 minutes of analysis. Using the values of the colorimetric coordinates, the chromatic tone angle (h°, 0°/360°=Red, 90°=yellow, 180°=green and 270°=blue) was calculated according to Equation 1, the Chroma (C*, color intensity or saturation that varies from gray to saturated color) according to Equation 2 (Wrolstad et al., 2005), and the global color difference (ΔE) according to Equation 3 (Pourjavaher et al., 2017).

$$h° = arctan(b*/a*) \quad (1)$$

$$C* = \sqrt{a*^2 + b*^2} \quad (2)$$

$$\Delta E = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2} \quad (3)$$

in which ΔL represents the variation of the coordinate L* ($\Delta L^* = L^* - L_0^*$), Δa represents the variation of the coordinate a* ($\Delta a = a^* - a_0^*$), e Δb represents the variation of the coordinate b* ($\Delta b = b^* - b_0^*$). L*, a* e b* represents the colorimetric coordinates of the films after exposure to acetic acid/ammonia vapors and L_0^*, a_0^* e b_0^* represents the colorimetric coordinates of the films before exposure to volatile compounds.

The detection limit (LOD) was considered the shortest exposure time of the films to acidic and basic vapors which the overall color difference was above 3.5 (ΔE > 3.5), a value considered visually noticeable by experienced and inexperienced observers (Fernández-Ramos et al., 2023; Mokrzycki & Tatol, 2011).

2.2.10 Thickness and mechanical properties

The film thickness (mm) was determined by a digital micrometer with an accuracy of 0.001 mm (Mitutoyo Corporation, Japan). The measurements were performed in 10 random points for each sample, with three repetitions for each treatment. The ultimate tensile strength (TS, MPa) and elongation at break (EAB, mm) were determined using the Universal Mechanical Testing Machine (Instron Corporation, Norwood, MA, USA) according to ASTM, D882-12 (ASTM, 2012). The samples (10 × 2.5 cm²) were fixed by two claws, with traction speed of 500 mm.min^{-1}. The results were expressed as mean of three repetitions and five replicates per treatment.

2.2.11 Statistical Analysis

Statistical models representing the influence of the factors on the color and the colorimetric change properties were performed using analysis of variance (ANOVA) (α = 0.05), with Tukey's Test or regression analysis. The model fitting was verified by the adjusted determination coefficient (R^2) and Akaike Information Criterion (AIC).

The linear models studied in this work used the adjusted coefficient of determination that weights the number of parameters, thus preventing models with more parameters from presenting a higher value of R^2_{adj} without actually explaining more about the phenomenon in question (Burnham & Anderson, 2000). Its expression is given by:

$$R^2_{adj} = 1 - ((1 - R^2)(n - 1)/(n - p)) \quad (4)$$

in which R^2 is the coefficient of determination, n is the number of observations and p is the number of the model parameters.

In nonlinear models, R^2 is referred to as the pseudo coefficient of determination because it does not have the same interpretation as R^2_{adj} does in linear models (Spiess & Neumeyer, 2010). The AIC is based on decision theory, admits the existence of a real model that describes the data and is unknown, and tries to select the model that comes closest to this "real model" (Burnham & Anderson, 2000). The AIC is calculated by the following expression:

$$AIC = -2ln\big(L(\Theta)\big) + 2p \quad (5)$$

in which ln(L(Θ)) is value of the natural base logarithm of the maximum point of the likelihood function and p is number of model parameters.

3. Results and Discussion

3.1 Bioactive compounds from blueberry extracts

The content of phenolic compounds and anthocyanins determined for blueberry extracts can be seen in Table 1.

Table 1 - Phenolic compounds and anthocyanin content of blueberry extracts.

Blueberry extracts	Total phenolic compounds (mg AGE/100g blueberries)	Total anthocyanins (mg cy-3-glu/100g blueberry)
Crude Extract (EB)	1392.35 ± 233.65 a	251.82 ± 14.27 a
Anthocyanin Extract (EFA)	900.55 ± 240.13 a	243.49 ± 19.94 a
Phenolic Extract (EF)	284.47 ± 51.86 b	ND

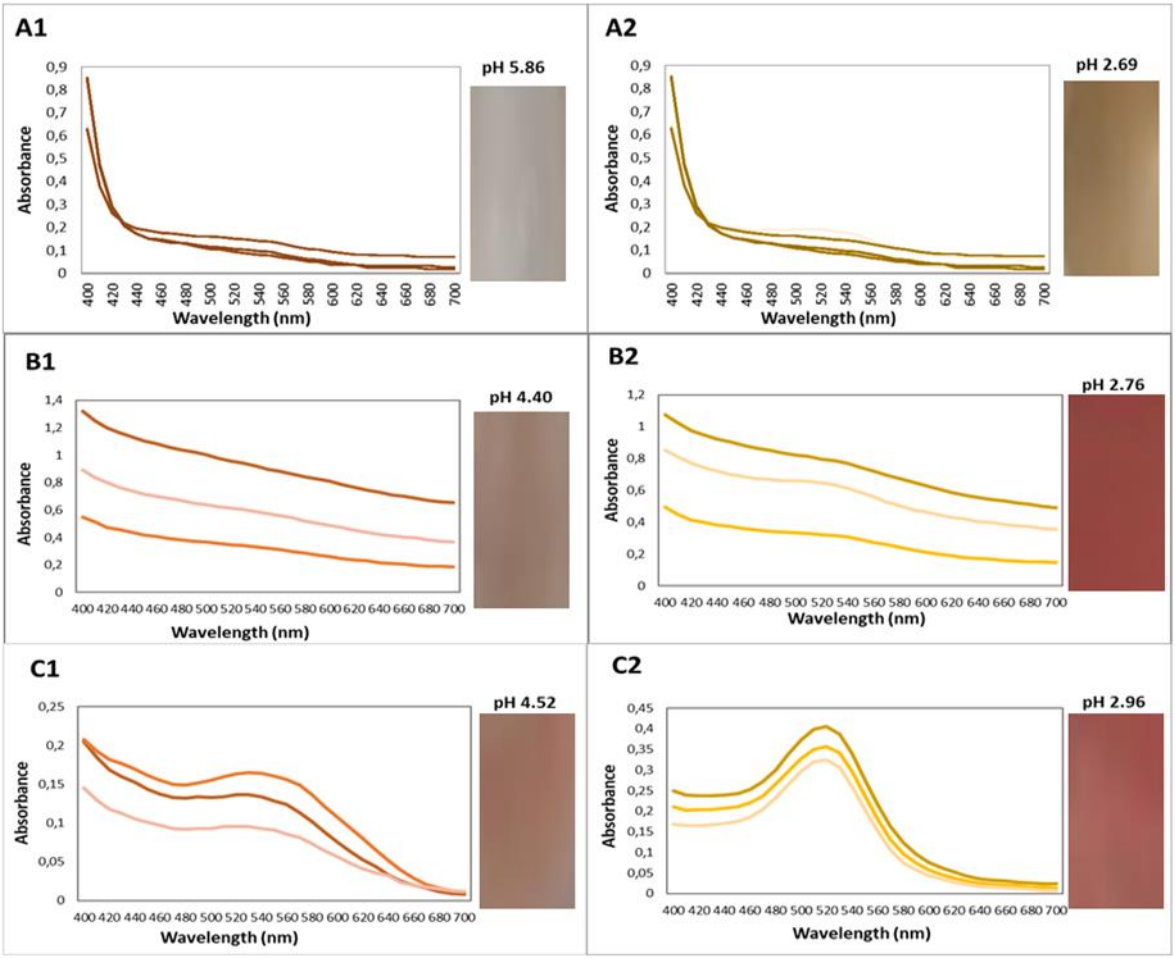
Data are expressed as mean ± standard deviation. AGE: gallic acid equivalent; cy-3-glu: cyanidin-3-glucoside. ND: Not detected. Means followed by the same letter, in the column, do not differ statistically using the Tukey and Student's t test at a significance level of 5%. Source: Prepared by the authors.

The EB and the EFA exhibited higher concentrations of total phenolic compounds and total anthocyanins compared to the EF sample ($p < 0.05$). This can be attributed to the prevalence of anthocyanins as the primary phenolic compounds in blueberries, followed by phenolic acids (Bai et al., 2023). Contents of total phenolic compounds and total anthocyanins can vary depending on differences in extraction methodology (types of solvent), the raw material used (blueberry pomace or the whole fruit), and also due to variation in harvests, location where the fruit is obtained, and environmental and experimental conditions. For example, Linhares et al. (2018) reported a total phenolic compound content of 420 mg of AGE and 44.26 mg of Cy-3-Glu in 100 g, after extraction from dry blueberry pomace using methanol/water (50:50, v/v) and acetone/water (70:30 v/v) as solvents. On the other hand, dos Santos et al. (2021) found 502 mg of GAE and 59 mg of Cy-3-Glu in 100 g of blueberry aqueous extract, obtained through extraction assisted by ultrasound bath (40 kHz and 120 W for 45 min).

3.2 Colorimetric sensitivity of blueberry extracts under exposure to acidic vapor

UV-Vis spectroscopy was used to explore the absorption of electromagnetic radiation, in the visible region, by components of blueberry extracts (EB, EFA, and EF), before and after exposure to acid vapor for 1 h. The obtained spectra are shown in Figure 1.

Figure 1 - UV-Vis spectrum of blueberry extracts before exposure to acid vapor: Phenolic extract - EF (A1); Crude extract - EB (B1) and Anthocyanin extract - EFA (C1). UV-Vis spectrum of blueberry extracts after exposure to acid vapor: Phenolic extract (A2); Crude extract (B2) and Anthocyanin extract (C2). The three lines represented by a different color within each of the six graphs represent the three replications of the experiment.



Before exposure to acid vapors, EB and EFA presented a pH around 4 and 5, in which the anthocyanins are mostly in the colorless form of pseudo-carbinol base (hemiketal form), that is, there is an interaction of flavylium cations and hydroxyl ions. Due to the presence of the flavylium cation, EB and EFA exhibited a less intense red color (Figure 1B1 and 1C1). When exposed to acid vapors, the pH of the extracts decreased to values below 3, similar to the outcomes reported by Fei et al. (2021). Consequently, in this condition, the majority of the anthocyanin exist in the form of flavylium cations (Zeng et al., 2019), exhibiting a more intense red color, as shown in the Figures 1B2 and 1C2. Therefore, an increase in the color intensity of EB and EFA was observed after 60 min of exposure to acid vapors.

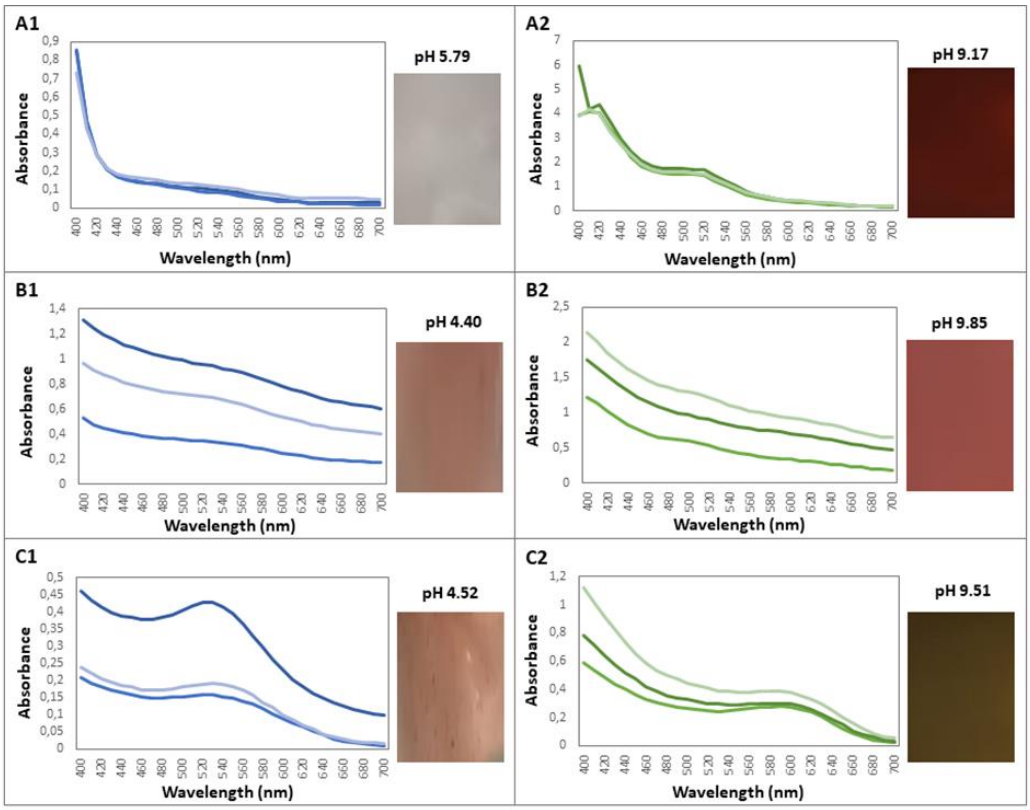
A hypsochromic (shift to a shorter wavelength) and hyperchromic (increase in the intensity) shift of the peak of maximum absorption was observed after exposing EFA to acid vapors (Figure 1C2). The absorption bands in the range of 500 nm to 560 nm and maximum peak at 520 nm were attributed to the flavylium cation structure characteristic of anthocyanins at pH 2.96 (Kamer et al., 2022; Zeng et al., 2019), which are present in high concentration and purified in EFA.

On the contrary to the observed for EF, an absorption band in the range of 500 nm to 560 nm was verified for EB sample, which can be attributed to the presence of anthocyanin. However, this band did not appear as well defined in the EB as in the EFA spectrum, possibly explained by the presence of other interfering phenolic compounds. Thus, a more orange appearance of EB was observed due to the yellow pigmentation of these compounds blended with the reddish color of the anthocyanin. Finally, in the EF and EB spectra, a pronounced absorption at 400 nm was observed, attributed to the yellow phenolic compounds in both extracts (Figure 1A1, 1A2, 1B1, and 1B2). The results suggested that acidic vapors had a greater impact on the chromatic behavior of anthocyanin compared to other phenolic compounds present in the blueberry extract.

3.3 Colorimetric sensitivity of blueberry extracts after exposure to volatile bases

Similarly, UV-Vis spectroscopy was used to explore the absorption of electromagnetic radiation by blueberry extracts before and after exposure to volatile bases. The spectra can be visualized in Figure 2.

Figure 2 - UV-Vis spectrum of blueberry extracts before exposure to ammonia vapor: Phenolic extract - EF (A1); Crude extract - EB (B1) and Anthocyanin extract - EFA (C1). UV-Vis spectrum of blueberry extracts after exposure to ammonia vapors: Phenolic extract (A2); Crude extract (B2) and Anthocyanin extract (C2).



Ammonia vapor was absorbed by the crude, phenolic, and anthocyanin extracts, increasing the pH values, corroborating to the study carried out by Fei et al. (2021). The carbinol pseudo-base form of anthocyanin present in EB and EFA prevails in pH 4.40 and pH 4.52, respectively (Neuenfeldt et al., 2022). With increasing pH, the number of conjugated double bonds decreases (Tarone et al., 2020) and, in alkaline conditions (pH > 8.0), anthocyanins are predominantly in the form of a quinoidal base in solution (Neuenfeldt et al., 2022).

The behavior observed for EB and EFA (Figure 2B2 and 2C2) possibly occurred due to the weakly neutralization of the NH_3 vapor dissolved in the aqueous medium by the free COOH groups of the anthocyanidin, causing a rapid increase in pH. This allowed the loss of protons from the anthocyanidin nucleus and the concomitant formation of other colored forms, that is, the neutral and anionic quinoid bases of violet and blue colors, respectively (Fei et al., 2021).

The EFA absorption band (520 nm), after exposure to ammonia vapor, underwent a bathochromic (610 nm) and hypochromic shift, due to the increase in the pH value of the solution from 4.52 to 9.51 (Fei et al., 2021; Freitas et al., 2020). The color change from pink to

green and the shift in the maximum absorption band were related to the transformation of the chemical structure of anthocyanins (Zeng et al., 2019), showing the predominance of the carbinol pseudo-base at pH 4.52 (Figure 2B1), and the quinoidal base when in pH 9.51 (Kamer et al., 2022). These color transformations corroborate the data reported by Kamer et al. (2022), Teixeira, Oliveira, et al. (2022) and Zeng et al. (2019).

The color of EF changed from light yellow to rusty brown when the pH increased from 5.79 to 9.17. In the EF absorption spectrum, the formation of a new band was observed at wavelengths between 400-420 nm and between 500-540 nm, due to the presence of phenolic compounds, which turned brown in an alkaline environment.

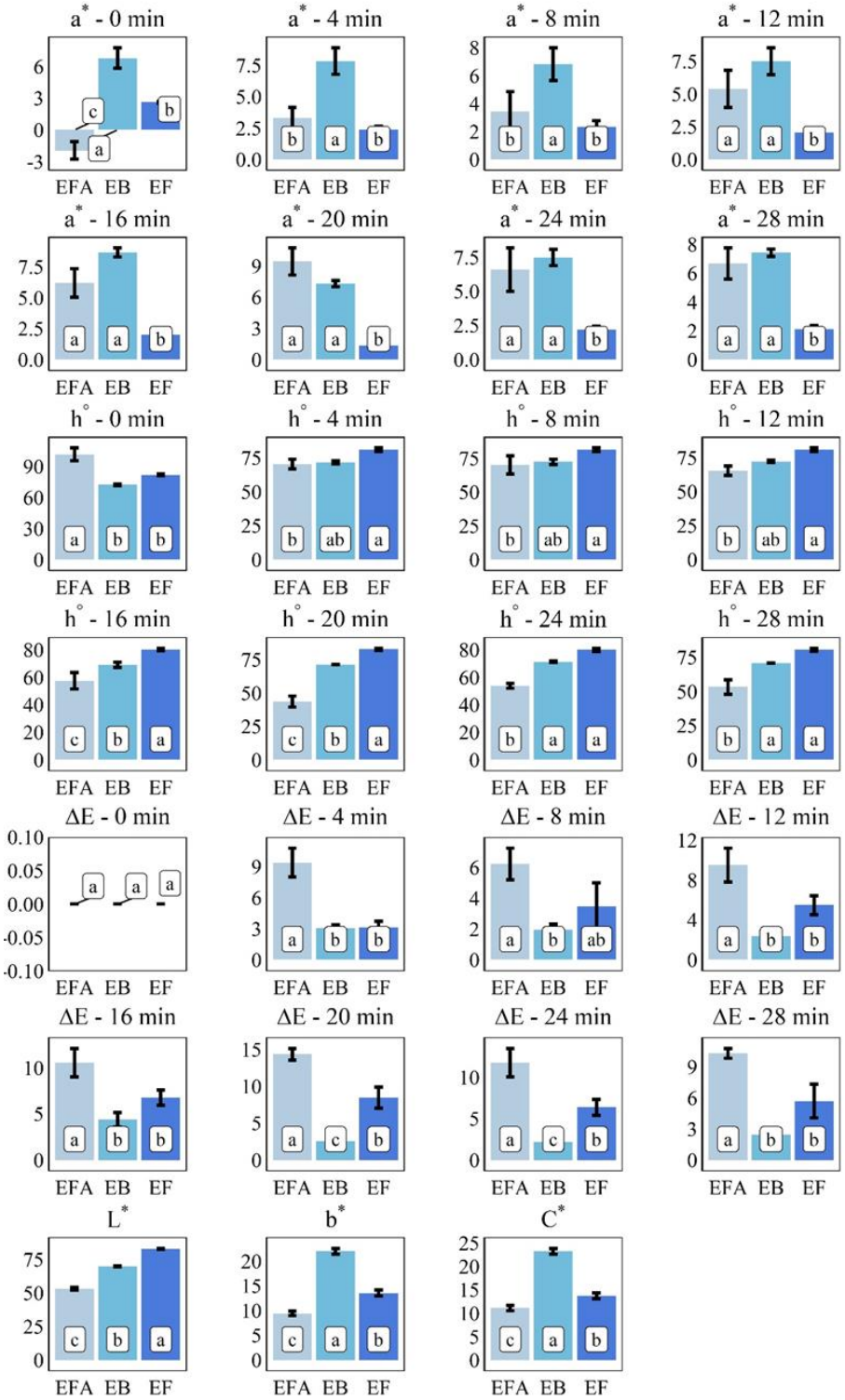
Well-defined absorption bands and significant band shift effects were not observed in the EB absorption spectrum. This behavior was probably due to the interaction of anthocyanins with other compounds, such as phenolic compounds, which stabilized the structural form of the anthocyanin as a function of pH. However, as the pH increased from 4.40 to 9.85, the color of the EB was intensified (Figure 2B1 and 2B2), attributed to the hypsochromic effect.

The color change of the solutions was measured to validate their potential use as indicators sensitive to changes in pH (Zeng et al., 2019). Indeed, there were visible changes from light pink to dark pink (EB), from light pink to dark green (EFA), and from light yellow to rust brown color (EF). Therefore, the color of blueberry extract solutions showed variations perceptible to the naked eye after exposure to volatile ammonia, showing potential for use as colorimetric indicators.

3.4 Colorimetric sensitivity of films after exposure to volatile acids

The colorimetric analysis of films incorporated with EF, EB, and EFA was used to explore the color change in films during exposure to volatile acids. The type of extract (EF, EB, and EFA) and the analysis time (0, 4, 8, 12, 16, 20, 24, and 28 min) significantly influenced the colorimetric coordinates and color attributes of the films, after exposure to acidic vapors, as can be seen in Figure 3.

Figure 3 - Mean values and standard deviation of the color attributes a*, hº, ΔE, L*, b* and C* of the films depending on the type of extract added and the time of exposure to acid vapors.



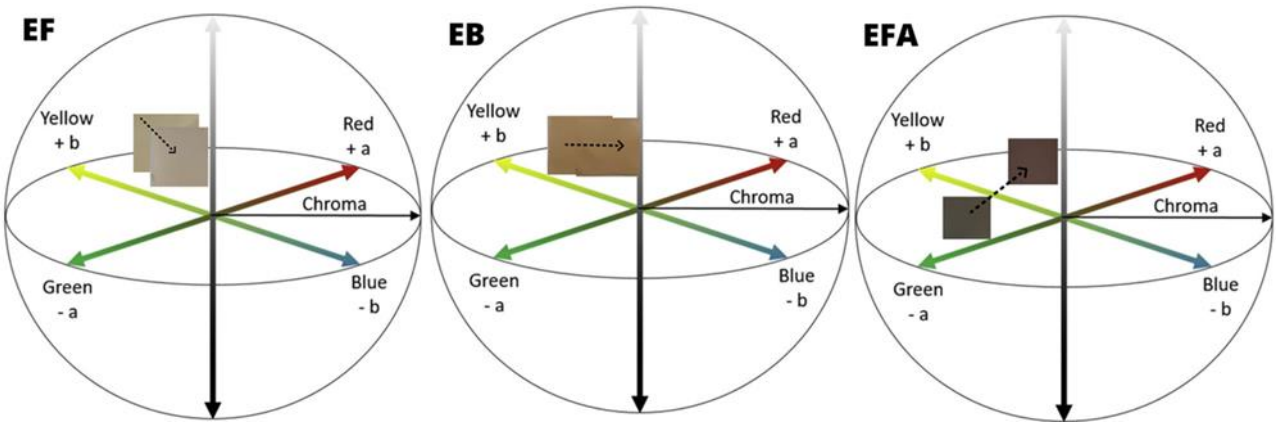
Pairs of means followed by at least the same lowercase letter do not differ from each other, at 5% significance, using the Tukey test. a*: Chromatic coordinate; h°: Chromatic tone angle; ΔE: Global color difference; L*: Brightness; b*: Chromatic coordinate; C*: Chroma. EB: Crude blueberry extracts; EF: Phenolic extract and EFA: Anthocyanin extract.

The color coordinates L*, b* and C* were not affected by the factor time ($p \geq 0.05$), however, they were affected by the type of extract used. The films with EF could be considered the lightest ones (highest L*) while the samples containing EFA were the darkest (lowest L*). As for the films with EB, a greater tendency to yellow color (higher positive b* value) and greater color intensity (higher C*) were observed, followed by the film with EF.

For the other colorimetric coordinates and attributes (a*, h^o, and ΔE), the interaction between the factors time and type of extract was significant ($p < 0.05$). The h^o results obtained for films containing EFA indicated that this was the only treatment studied to show a significant difference in tone over time, which was already noticeable in the first 4 minutes of exposure, becoming more evident over time. After 28 min, the h^o attribute varied from 101.53º (greenish color), before exposure to acid vapor, to 53.11º, exhibiting a purple color (Figure 4). This chromatic transition property is important when applying these films as colorimetric indicators, sensitive to changes in pH resultant from the presence of acid vapors. Also, the film incorporated with EFA shown a significant difference over time for the a* coordinate, starting with a negative value (-1.96, green), to a positive value (6.65, red), reinforcing the color changing in this film under the conditions studied.

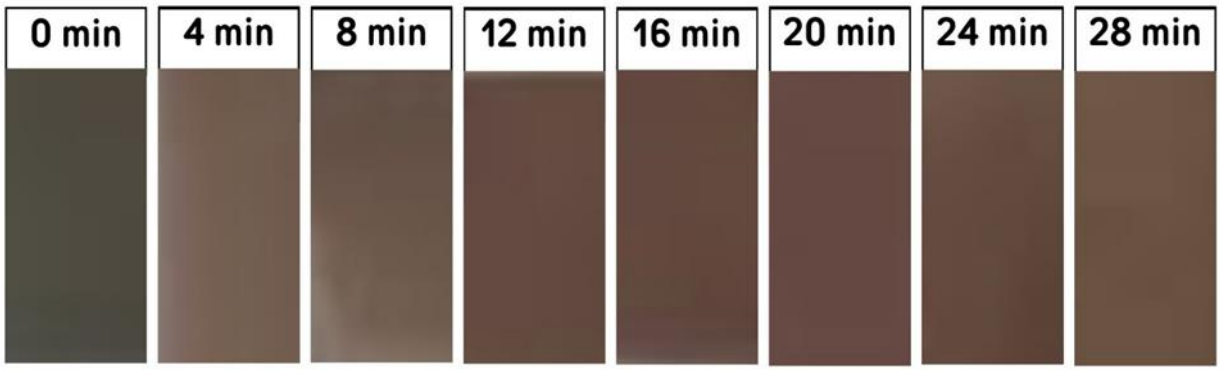
Concerning the overall color difference (ΔE), values higher than 3.5 are interesting since this difference is noticeable by experienced and inexperienced observers, i.e., samples showing this behavior are promising as colorimetric indicators in intelligent packaging. In this sense, the films with EFA presented the highest ΔE (10.31) compared to the other films, after 28 min of exposure to acid vapors (Figure 3). On the other hand, in terms of colorimetric sensors, low values of ΔE are not desirable because they indicate subtle or non-perceptible color changes (Silva; et al., 2013), as occurred for the films with EB. The film with EF showed a significant difference over time ($p < 0.05$), however the values were lower than the film added with EFA, after 28 min of exposure. The data corroborated the color behavior observed visually in the films, as shown in Figure 4.

Figure 4 - Chromatic behavior of PVA and gelatin films with blueberry extracts, after exposure to acid vapor, in the CIELAB color space. EF: Phenolic extract. EB: Crude extract. EFA: Anthocyanin extract. Start of the arrow refers to time zero and end of the arrow refers to the 28 min exposure time.



The films added with EFA showed a rapid color change (green to purple), in a response time of 4 min (ΔE = 9.39) (Figure 3 and 5). Therefore, the LOD for these films occurred before 4 min (Figure 5), presenting the lowest LOD among the samples studied. The films with EF reached ΔE > 3.5 around 8 and 12 min of exposure (ΔE = 5.47 at 12 min), and the film with EB, only after 16 min (ΔE = 4.37).

Figure 5 - Colorimetric response of the film with EFA after exposure to volatile acid for 28 min.



In this context, the color of the film with blueberry EFA showed noticeable variations to the naked eye, after exposure to the volatile acid, showing potential for use as a colorimetric indicator film in these conditions.

The statistical models that represent the influence of time of exposure to volatile acids in PVA/gelatin films with different fractions of blueberry extract, for the color coordinates L*, a*,

b*, h°, C*, and ΔE, as well as the values of R^2_{adj} and AIC, are summarized in Table 2 and Figure 6.

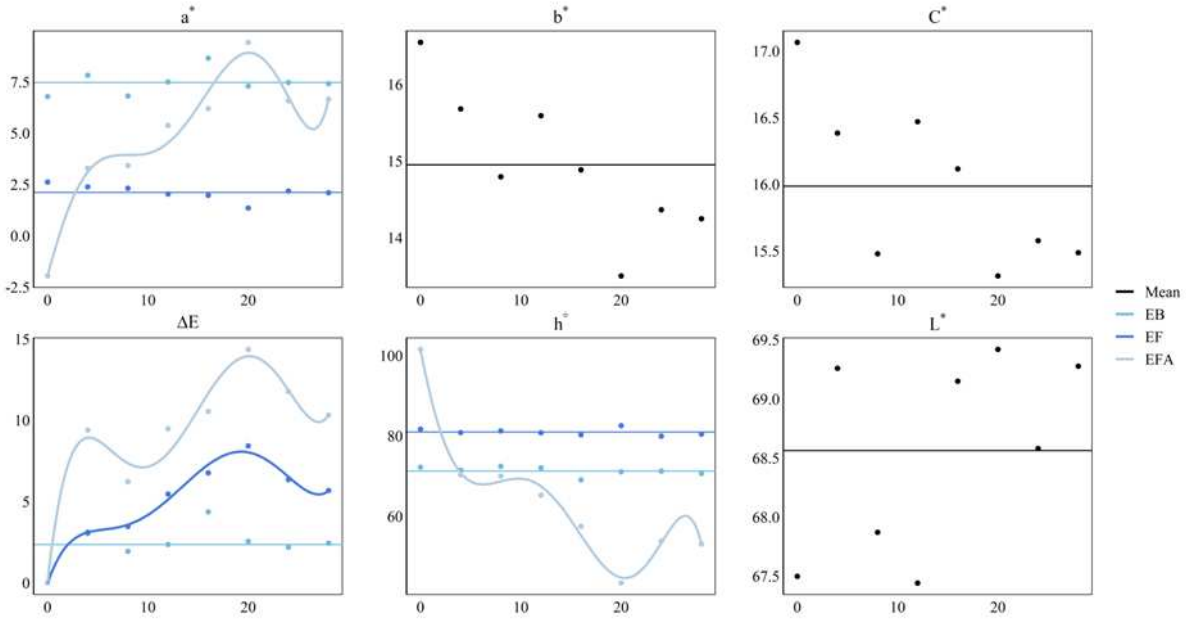
Table 2 - Linear models, R^2_{adj}, and AIC of the color coordinates (L*, a*, b*, ΔE, h°, and C*) for the films incorporated with crude (EB), phenolic (EF) and anthocyanin (EFA) extracts over time of exposure to acidic vapors.

Color coordinates / Extract type (E)	Linear Models	R^2_{adj}	AIC
L*	$y = 68.5626 \pm 14.83$	-	-
a*			
EF	$y = 2.1175 \pm 0.37$	-	-
EB	$y = 7.4675 \pm 0.59$	-	-
EFA	$y_i = -1.9358 + 1.8341x_i - 0.0838x_i^2 - 0.0268x_i^3 + 0.0036x_i^4 - 0.0001x_i^5 + 0.000002x_i^6$	0.8255	27.6057
b*	$y = 14.9507 \pm 6.41$	-	-
C*	$y = 15.9865 \pm 6.39$	-	-
h°			
EF	$y = 80.9633 \pm 0.83$	-	-
EB	$y = 71.2809 \pm 1.07$	-	-
EFA	$y_i = 101.4678 - 12.8743x_i + 1.2791x_i^2 + 0.0507x_i^3 - 0.0139x_i^4 + 0.0006x_i^5 - 0.000009x_i^6$	0.9546	43.2413
ΔE			
EF	$y_i = -0.0040 + 1.8139x_i - 0.3879x_i^2 + 0.0381x_i^3 - 0.0015x_i^4 + 0.00002x_i^5$	0.9586	15.5862
EB	$y = 2.3634 \pm 1.21$	-	-
EFA	$y_i = 0.0766 + 5.6406x_i - 1.2315x_i^2 + 0.1091x_i^3 - 0.0040x_i^4 + 0.00005x_i^5$	0.8613	33.1623

L*: Luminosity; a*, b*: Chromatic coordinate; ΔE: Global color difference; h°: Chromatic tone angle; C*: Chroma; x: Exposure time. The equations adjusted were significant ($p < 0.05$) by ANOVA. Source: Prepared by the authors.

High coefficient of determination (R^2) values indicated that the variable under analysis followed a pattern, thus being possible to fit a model. The regression models that better fitted the results were linear models, presenting the highest R^2 and lowest AIC values (Table 2), which are also presented in the form of graphs (Figure 6).

Figure 6 - Linear Models, R^2_{adj}, AIC or mean of the color attributes (L*, a*, b*, ΔE, h° and C*) of the films incorporated with crude (EB), phenolic (EF) and anthocyanin (EFA) extracts after exposure for 28 min to acid vapors.



L*: Brightness; a*, b*: Chromatic coordinate; ΔE: Global color difference; h°: Chromatic tone angle; C*: Chroma.

It was possible to predict the behavior of the color coordinates of films, when exposed to acid vapor, since a pattern was observed over time. Figure 6 evidences that the regression graphs presented more than one maximum and one minimum, a behavior similar to that of linear functions, confirming that the polynomial functions of degree 5 and 6 better explained the color behavior of the films after the exposure to acid vapor for 28 min.

3.5 Colorimetric sensitivity of films after exposure to volatile bases

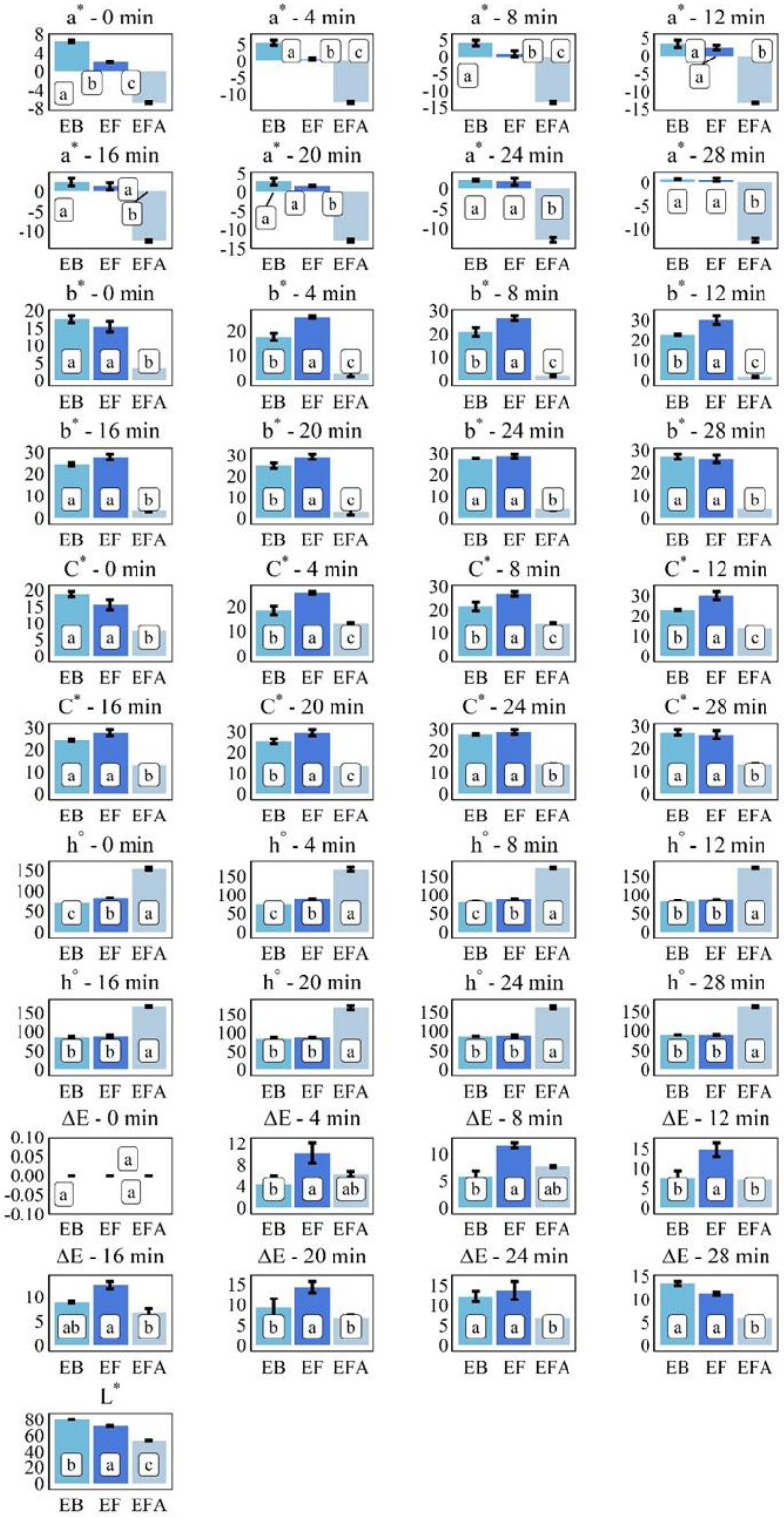
Volatile nitrogen components are often generated as a result of microbial spoilage in protein-rich foods (Kamer et al., 2022). The gas NH_3 is one of the most important spoilage gases released during meat, fish, and seafood storage period (Fei et al., 2021). Due to the importance of this compound, the presence of ammonia vapor that can be released due to the degradation of foods with high-protein content was simulated and the response of the color indicator films was determined. Figure 7 shows the variation in color coordinates of films (L*, a*, b*, C*, hº, and ΔE) stored in the presence of ammonia for 28 minutes.

The increase in b* coordinate occurred in EF and EB films due to the yellow color of the films. This effect corroborates the chromatic behavior of the respective extract solutions (Figure

2), attributed to the presence of yellow non-anthocyanin phenolic compounds in the extracts such as flavonols (e.g., myrcetin and quercetin) (Pires et al., 2020), and phenolic acids (e.g., caffeic acid and ferulic acid) (Bai et al., 2023), which can be present.

The interaction between the factors time and type of extract was non-significant ($p \geq 0.05$) for L*, however, both factors were significant when studied individually ($p < 0.05$). The films developed with EF were the brightest (highest L*), while the EFA was the darkest (lowest L*). However, all films became darker after 28 min of exposure to ammonia (Figure 7). For the other color coordinates (a*, b*, C*, h^o, and ΔE), the interaction between the factors time and type of extract was significant ($p < 0.05$). It was observed that, for all films, there was an increase in C* after exposure to ammonia, with an increase of color intensity of the films over time. Among the films tested, EFA film was the one that presented the smallest C* values.

Figure 7 - Mean values and standard deviation of the color attributes L*, a*, b*, C*, hº and ΔE of the films depending on the type of extract added and the time of exposure to ammonia.



Pairs of means followed by at least the same lowercase letter do not differ from each other, at 5% significance, using the Tukey test. a*, b*: Chromatic coordinate; C*: Chroma; h°: Chromatic tone angle; ΔE: Global color difference; L*: Brightness. EB: Raw blueberry extracts; EF: Phenolic extract and EFA: Anthocyanin extract.

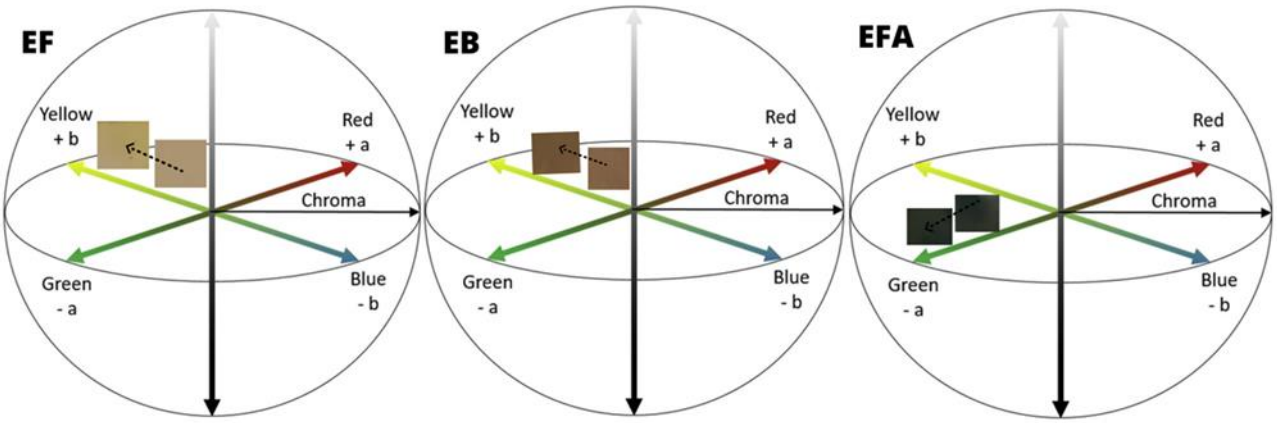
On the contrary to the other film treatments, the EB film showed a significant difference in the chromatic tone angle, compared to the initial time (0 min) and the final time (28 min), from 69.52º to 88.45º, demonstrating the sensitivity of this material to pH changes resulting from basic vapors. The increase in the shade angle is due to the yellow color approaching and the red color moving away with time of exposure to ammonia.

The value of the a* coordinate of the EB and EFA films reduced with the time of exposure to ammonia, corroborating the study carried out by Aydogdu Emir (2023). This reduction can be explained by the structural alteration of anthocyanins present in blueberry extracts (De Souza et al., 2015; Idham et al., 2012). The EFA films showed a color closer to green (a* negative), similar to the color of pure EFA after exposure to ammonia (pH 9.0) (Figure 2).

The ΔE of the films increased after exposure to ammonia vapor. However, in contrast to what was reported when the films were exposed to acid vapors, ΔE for EFA films was lower than the ΔE verified for EF and EB films, after 28 min of exposure. Therefore, when the films were exposed to basic vapors, the color change was more noticeable in these last two treatments.

After 4 minutes of exposure to the basic vapor, the three film treatments presented $\Delta E > 3.5$ (EFA: 6.25; EF: 10.12; EB: 4.29), indicating that the LOD for all films occurred before 4 min of exposure. Furthermore, the ΔE values were significantly ($p < 0.05$) different and gradually increased with time, mainly in the EB film. In films with EF and EFA, there was no significant difference in ΔE ($p \geq 0.05$) after 4 min of exposure, therefore, given the color pattern obtained in the 4 min time, the colors of the other times are not distinguishable. The data corroborated the chromatic behavior of the films (Figure 8).

Figure 8 - Chromatic behavior in CIELBA space of PVA and gelatin films added with blueberry extracts after exposure to ammonia vapor. EF: Phenolic extract. EB: Crude extract. EFA: Anthocyanin extract. Start of the arrow refers to time zero and end of the arrow refers to the 28 min exposure time.



The colorimetric changes in PVA/gelatin films are based on the principle that vapor ammonia has penetrated the film structure, creating alkaline conditions (Kamer et al., 2022). The interaction between the NH_3 molecules and the hydroxyl groups present on the surface and inserted between the polymer chains of the film produced NH_4^+ and OH^- species. This process induced changes in the chemical structures of the anthocyanins present in the film added with EB and EFA, consequently modifying the color attributes of the films (Kamer et al., 2022).

It was possible to fit linear and non-linear models (Table 3 and Table 4, respectively) to the color coordinates, according to the two-away ANOVA interpretation for the PVA/gelatin-based films incorporated with different types of blueberries extracts after exposure to volatile bases. The interaction time x type of extract was not significant ($p \geq 0.05$) for L* and this coordinate was only affected by time individually ($p < 0.05$). Therefore, since the type of extract used did not influence the L*, only one linear and one non-linear models were presented. For C* and ΔE, the interaction was significant ($p < 0.05$). In addition, the coordinates a* and h^o (for EF) and b* (EFA) were not influenced by the factor time, being thus represented by the mean and standard deviation in Tables 3 and 4.

Table 3 - Linear models, R^2_{adj}, and AIC of the color coordinates (L*, a*, b*, ΔE, h°, and C*) for the films incorporated with crude (EB), phenolic (EF) and anthocyanin (EFA) extracts after exposure for 28 min to volatile bases.

Color coordinates / Extract type (E)	Linear Models	R^2_{adj}	AIC
L*	$y_i = 70.4180 - 0.4576x_i + 0.0320x_i^2 - 0.0007x_i^3$	0.9682	2.6216
a*			
EF	$y_i = 1.3550 \pm 0.63$	-	-
EB	$y_i = 6.4653 - 0.1247x_i - 0.0427x_i^2 + 0.0032x_i^3 - 0.00006x_i^4$	0.9770	6.7690
EFA	$y_i = -6.8463 - 2.2066x_i + 0.2432x_i^2 - 0.0105x_i^3 + 0.0001x_i^4$	0.9834	6.5244
b*			
EF	$y_i = 15.4723 + 3.4582x_i - 0.3412x_i^2 + 0.0147x_i^3 - 0.0002x_i^4$	0.9251	30.6149
EB	$y_i = 17.4711 - 1.2272x_i + 0.4564x_i^2 - 0.0435x_i^3 + 0.0017x_i^4 - 0.00002x_i^5$	0.9931	7.6446
EFA	$y = 2.9958 \pm 0.84$	-	-
C*			
EF	$y_i = 15.5877 + 3.4233x_i - 0.3364x_i^2 + 0.0144x_i^3 - 0.0002x_i^4$	0.9233	30.7534
EB	$y_i = 18.6495 - 1.2766x_i + 0.4408x_i^2 - 0.0416x_i^3 + 0.0016x_i^4 - 0.00002x_i^5$	0.9934	5.5847
EFA	$y_i = 7.6744 + 2.2026x_i - 0.2789x_i^2 + 0.0149x_i^3 - 0.0003x_i^4 + 0.000002x_i^5$	0.9884	1.0725
h°			
EF	$y = 86.7789 \pm 1.97$	-	-
EB	$y_i = 69.5005 + 0.0592x_i + 0.2938x_i^2 - 0.0265x_i^3 + 0.0008x_i^4 - 0.000009x_i^5$	0.9842	22.5222
EFA	$y_i = 153.1291 + 5.2885x_i - 0.4881x_i^2 + 0.0168x_i^3 - 0.0002x_i^4$	0.8647	40.1385
ΔE			
EF	$y_i = 0.1101 + 3.5409x_i - 0.3516x_i^2 + 0.0151x_i^3 - 0.0002x_i^4$	0.9297	30.4063
EB	$y_i = -0.0041 + 2.6787x_i - 0.6929x_i^2 + 0.0963x_i^3 - 0.0065x_i^4 + 0.0002x_i^5 - 0.000002x_i^6$	0.9969	-0.5807
EFA	$y_i = 0.0124 + 2.4632x_i - 0.2718x_i^2 + 0.0118x_i^3 - 0.0001x_i^4$	0.9969	-5.0993

L*: Luminosity; a*, b*: Chromatic coordinate; ΔE: Global color difference; h°: Chromatic tone angle; C*: Chroma; x: Exposure time. The equations adjusted were significant ($p < 0.05$) by ANOVA. Source: Prepared by the authors.

Table 4 - Non-linear models, R², and AIC of the color coordinates (L*, a*, b*, ΔE, h° and C*) for the films incorporated with crude (EB), phenolic (EF) and anthocyanin (EFA) extracts after exposure for 28 min to volatile bases.

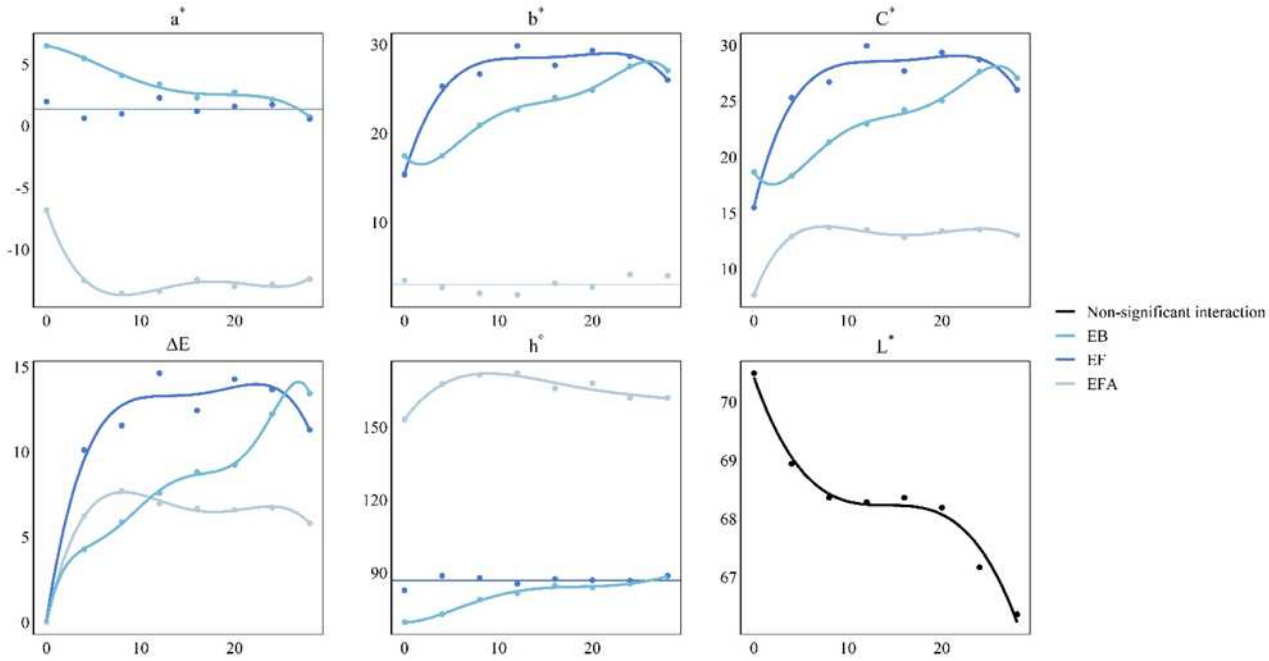
Color coordinates/ Extract type (E)	Non-linear Models	R^2	AIC
L*	$y_i = 69.8716e^{-0.0016x_i}$	0.8467	15.6732
a*			
EF	$y_i = 1.3550 \pm 0.63$	-	-
EB	$y_i = \frac{6.50 + 0.0097x_i^2}{0.0103x_i^2 + 1}$	0.9379	15.4724
EFA	$y_i = -5 - \frac{19.4767x_i^2}{2.4659x_i^2 + 1}$	0.8629	24.1741
b*			
EF	$y_i = 15 + \frac{2.8645x_i^2}{0.2148x_i^2 + 1}$	0.9250	31.3943
EB	$y_i = 17.7928e^{0.0167x_i}$	0.9337	27.7216
EFA	$y = 2.9958 \pm 0.84$	-	-
C*			
EF	$y_i = 15 + \frac{2.8442x_i^2}{0.2123x_i^2 + 1}$	0.9211	31.7585
EB	$y_i = 18.6076e^{0.0149x_i}$	0.9374	25.6374
EFA	$y_i = 7 + \frac{7.5618x_i^2}{1.1928x_i^2 + 1}$	0.9618	12.6378
h°			
EF	$y = 86.7789 \pm 1.97$	-	-
EB	$y_i = 72.1584e^{0.0077x_i}$	0.8954	39.6443
EFA	$y_i = 155 + \frac{0.4445x_i^2}{0.0001x_i^4 + 1}$	0.7299	46.4480
ΔE			
EF	$y_i = \frac{2.6144x_i^2}{0.1962x_i^2 + 1}$	0.9402	29.8833
EB	$y_i = 0.7920x_ie^{-0.0196x_i}$	0.9625	24.7868
EFA	$y_i = \frac{14.0476x_i^2}{2.0913x_i^2 + 1}$	0.9502	17.7666

L*: Luminosity; a*, b*: Chromatic coordinate; ΔE: Global color difference; h°: Chromatic tone angle; C*: Chroma; x: Exposure time. The equations adjusted were significant ($p < 0.05$) by ANOVA. Source: Prepared by the authors.

The linear regression models, of degrees 3, 4, 5 and 6 (Table 3), and the non-linear regression models, expressed by exponential functions and $ax^2/(bx^2+1)$ (Table 4), were the ones that best adjusted to the color coordinates in films after 28 minutes of exposure to volatile bases, showing the highest values of R^2_{adj} and lowest values of AIC, as well as the highest values of R^2, respectively (Burnham & Anderson, 2000).

Observing the results in Tables 3 and 4, the ΔE over time of the EF film was better explained by the exponential function, since the smaller the AIC, the better the fit of the data to the model (Burnham & Anderson, 2000). For the other color parameters, the polynomial functions (linear model) best explained the color behavior of the films over the time of exposition to the basic vapor. The graphs of the best-fitting linear models are shown in Figure 9, as in most of the cases studied these models presented better goodness-of-fit measures.

Figure 9 - Linear Models, Radj², AIC of the color attributes (L*, a*, b*, ΔE, h° and C*) of the films incorporated with crude (EB), phenolic (EF) and anthocyanin (EFA) extracts after exposure for 28 min to volatile bases.



L*: Brightness; a*, b*: Chromatic coordinate; ΔE: Global color difference; h°: Chromatic tone angle; C*: Chroma.

3.6 Thickness and mechanical properties

The results obtained for thickness, tensile strength (UTS), and elongation at break (EAB) of the elaborated films are presented in Table 5. This analysis aimed to verify if the

incorporation of the extracts would compromise the PVA/gelatin films' mechanical performance. In this sense, for this test in particular, the comparisons were made considering a control film elaborated without any extract.

Table 5 - Thickness, tensile strength, and elongation at break determined for films.

Film	Thickness (mm)	Tensile strength (MPa)	Elongation at break (mm)
PVA/gelatin (control)	0.16 ± 0.007 a	5.22 ± 0.561 a	176.00 ± 20.435 a
PVA/gelatin with EB	0.17 ± 0.011 a	4.57 ± 0.367 ab	148.00 ± 11.029 b
PVA/gelatin with EFA	0.15 ± 0.003 a	4.21 ± 0.613 b	149.08 ± 9.923 b
PVA/gelatin with EF	0.15 ± 0.003 a	5.35 ± 0.485 a	132.58 ± 13.045 b

Data are expressed as mean ± standard deviation. Means followed by the same letter, in the column, do not differ statistically using the Tukey test at a significance level of 5%. Source: Prepared by the authors.

The thickness of the PVA/gelatin films, with or without blueberry extracts, were uniform ($p \geq 0.05$), with an average thickness of 0.16 mm. The values can be considered satisfactory for application as primary packaging and coating (Sganzerla et al., 2021). Similar results were reported by Sganzerla et al. (2021), when investigating carboxymethyl cellulose films incorporated with blackberry extract (*Morus nigra* L.).

Tensile strength was lower in films incorporated with EFA ($p < 0.05$). The anthocyanin molecules present in both extracts are large and, for this reason, provide a greater distance between the gelatin and PVA molecules, causing a reduction in the intra- and intermolecular interactions among the polymer chains (Musso et al., 2019). On the other hand, the incorporation of crude extract and purified fractions of phenolic extract did not jeopardize UTS in relation to the control film ($p \geq 0.05$). The elongation at break, however, decreased with the addition of blueberry extracts, that is, they induced a reduction in the flexibility of the PVA/gelatin polymeric blend. The incorporation of phenolic compounds can hinder chain-chain interactions in the films, making them more susceptible to tearing during stretching, which is a recurring problem in bio-based polymer films that deserves attention (Tymczewska et al., 2022).

The addition of anthocyanin extract also influenced the tensile strength and elongation at break of gelatin/polyvinyl alcohol films in the study by Zeng et al. (2019). The authors reported that the UTS value of the films decreased with increasing mulberry anthocyanins extract. Conversely, the elongation at break increased with the addition of the extract, different from

the results herein obtained. The addition of black cumin seed extract, rich in bioactive compounds, to the gelatin/PVA blend resulted in a slight decrease in UTS, as well as in EAB compared to the control film (without extract), as occurred in this study (Tymczewska et al., 2022).

4. Conclusion

The present study evaluated three blueberry extracts and their incorporation into polymeric films based on PVA/gelatin to develop indicators films. Although the extracts, when investigated individually, exhibited interesting chromatic transition, with high ΔE values when subjected to different pH conditions, the films obtained after their incorporation into the polymer matrix behaved differently. Films containing crude blueberry extract (EB) were the most sensitive to basic vapors, suggesting their suitability to monitor the shelf life of foods that release ammonia during deterioration. Additionally, the color change was clearly noticeable to the naked eye, which is an important advantage in terms of assisting the consumers when purchasing perishable foods. The findings also suggested that the films incorporated with anthocyanin extract (EFA) were the most sensitive to acidic vapors, indicating its potential to monitor de degradation of foods that acidify, such as milk. The blueberry phenolic extract (EF), on the other hand, proved not to be a good additive in terms of developing colorimetric indicator films since the films incorporated with this extract exhibited lower values of ΔE under the conditions tested. The films incorporated with EB and EFA, however, underperformed compared to PVA/gelatin films without any type of extract, mainly regarding the mechanical properties (i.e., elongation at break feature), indicating that their application into polymeric matrices must be assessed carefully. Finally, it is worth noting that the herein reported results were obtained through *in vitro* experiments. The evaluation of the films in real food systems is recommended in future studies to better assess the sensors' efficiency to monitor the food shelf life.

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Declaration of Conflict of Interest

There are no conflicts of interest to declare.

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Figure captions

Figure 1 - UV-Vis spectra of blueberry extracts before (1) and after (2) exposure to acid vapor: Phenolic extract - EF (A); Crude extract - EB (B) and Anthocyanin extract - EFA (C).

Figure 2 - UV-Vis spectrum of blueberry extracts before (1) and after (2) exposure to ammonia vapor: Phenolic extract - EF (A); Crude extract - EB (B) and Anthocyanin extract - EFA (C).

Figure 3 - Mean values and standard deviation of the color coordinates a* and b* (chromatic coordinate), hº (chromatic tone angle), ΔE (overall color difference), L* (luminosity), and C* (chroma) of the films depending on the type of extract added (raw blueberry extract 'EB', phenolic extract 'EF', and anthocyanin extract 'EFA') and the time of exposure to acid vapors (0, 4, 8, 12, 16, 20, 24, and 28 min). Pairs of means followed by at least the same lowercase letter do not differ from each other, at 5% significance, using the Tukey test.

Figure 4 - Chromatic behavior of PVA and gelatin films incorporated with blueberry extracts, after exposure to acid vapor, in the CIELAB color space. EF: Phenolic extract. EB: Crude extract. EFA: Anthocyanin extract. Start of the arrow refers to time zero and end of the arrow refers to 28 min of exposure.

Figure 5 - Color change in films with anthocyanin extract (EFA) during 28 min of exposure to volatile acid.

Figure 6 - Color coordinates L* (luminosity), a* and b* (chromatic coordinates), ΔE (overall color difference), h° (chromatic tone angle), and C*(chroma) of the films incorporated with crude (EB), phenolic (EF) and anthocyanin (EFA) extracts over time of exposure to acidic vapors (min).

Figure 7 - Means and standard deviation of the color coordinates L* (luminosity), a* and b* (chromatic coordinates), ΔE (overall color difference), h° (chromatic tone angle), and C*(chroma) of the films depending on the type of extract added (raw blueberry extracts 'EB', phenolic extract 'EF' and anthocyanin extract 'EFA') and the time of exposure to ammonia. Pairs of means followed by at least the same lowercase letter do not differ from each other, at 5% significance, using the Tukey test).

Figure 8 - Chromatic behavior in CIELAB space of PVA/gelatin films added with blueberry extracts after exposure to ammonia vapor. EF: Phenolic extract. EB: Crude extract. EFA: Anthocyanin extract. Start of the arrow refers to time zero and end of the arrow refers to the 28 min exposure time.

Figure 9 - Color coordinates L* (luminosity), a* and b* (chromatic coordinates), ΔE (overall color difference), h° (chromatic tone angle), and C*(chroma) of the films incorporated with

crude (EB), phenolic (EF) and anthocyanin (EFA) extracts over time of exposure to volatile bases.

4. ARTICLE 2: Improvement of food packaging properties of gelatin/polyvinyl alcohol films through blueberry extracts integration

Abstract

Films based on polyvinyl alcohol (PVA) and gelatin (GL) combined with different blueberry extracts, such as crude extract, phenolic extract, and anthocyanin extract, were prepared and characterized as a function of their thermal, physical, chemical, mechanical, and topological features. Thermal degradation of the films occurred in three stages. Moreover, the polymer blend of PVA and gelatin (FC) was the most thermal stable while the peak of maximum thermal degradation was higher for FB and FA. Regarding mechanical properties, despite the gelatin films (FG) that were the most rigid (highest Young's modulus - YM), all polymer blends exhibited greater flexibility (elongation capacity) and tensile strength due to the addition of PVA. Besides, the blueberry extracts incorporation did not influence the tensile strength at the break of the films ($p \geq 0.05$). On the other hand, high concentrations of anthocyanins performed as plasticizers, increasing the elasticity of FA, while FB and FF films were more rigid, and this effect can be attribute to lower concentrations of anthocyanins and the presence of other phenolic compounds. The FC exhibited an integrated, homogeneous, smooth surface with good dispersion and interaction of polymers, i.e., good compatibility between gelatin and PVA. A reduction in polymer dispersion was observed in FF and FB and a lower affinity was observed after the addition of blueberry anthocyanin extracts to the matrix, since a less homogeneous surface was observed in FA. In addition, the addition of blueberry extract hindered the penetration of water into the matrix, decreasing the swelling index of FA, FB and FF. Considering only the mixture and the pure polymers, FC was less soluble than FG and PVA (FPVA) films. Among the films with blueberry extract, FF was the least soluble in water, which can be attributed to the non-anthocyanic phenolic compounds, phenolic acids and flavonoids, present in the blueberry phenolic extract added to the FF, have free hydroxyls available to interact with the matrix of the GL-PVA-glycerol blend film to form hydrogen bonds. Therefore, the PVA, gelatin and blueberry extract films have potential to be applied as smart packaging for food. With emphasis on FB stood out because, in addition to presenting better physical, chemical and mechanical properties than FA, it presented better thermal stability than FF.

Keywords: Film characterization; Smart packaging; Purification; Alcoholic extracion; Natural extracts.

1. Introduction

Blueberries (*Vaccinium* spp.) are fruits rich in phenolic compounds such as stilbenoids, tannins, and flavonoid compounds, including anthocyanin, flavanone, flavanol, and quercetin (BAI et al., 2023; JARA et al., 2019; PIRES et al., 2020), with anthocyanin being the most abundant phenolic compound in these fruits, followed by phenolic acid (BAI et al., 2023). Due to their rich composition, blueberries have been nicknamed a "superfruit" by the Food and Agriculture Organization of the United Nations (FAO) and are considered one of the five healthiest fruits in the world (BAI et al., 2023).

Given the bioactive richness of blueberries, physicochemical processes are employed to obtain extracts rich in these compounds, which have broad applications in various fields, especially in the Food Industry. By using extraction with only a non-specific solvent, such as ethanol, an extract containing a wide range of substances is obtained, including sugars, amino acids, anthocyanins, phenolic compounds, flavonoids, and phenolic acids. Once concentrated, these substances form what is known as the crude blueberry extract, characterized by low purity (BAI et al., 2023; GOMES et al., 2024; RODRIGUEZ-SAONA; WROLSTAD, 2001b).

However, by employing a purification process with specific solvents, it is possible, after extraction, to separate the compounds present in the crude extract (RODRIGUEZ-SAONA; WROLSTAD, 2001b) and obtain two more purified extracts: the anthocyanin extract (EFA), in which the anthocyanins from blueberries are present in higher concentrations; and the phenolic extract (EF), which contains other phenolic compounds from blueberries, such as phenolic acids, flavonols, and flavan-3-ols, in higher concentrations as well (COKLAR; AKBULUT, 2017; GOMES et al., 2024; PIRES et al., 2020).

Some phenolic compounds are used as natural pigments and preservatives in several foods, such as sweets and snacks, targeting a consumer audience more concerned with healthiness. Currently, these substances are also being explored as components of polymeric films due to their antioxidant properties and their role as indicator dyes. When incorporated into polymeric matrices, they impart active and/or intelligent properties to these matrices, contributing to the extension of the packaged product's shelf life or enabling the monitoring of its quality during storage (BAI et al., 2023; MA; DU; WANG, 2017; YONG et al., 2019a).

In this sense, various biopolymers have been used to develop films with a more sustainable appeal and active and/or intelligent properties. Among them, polyvinyl alcohol (PVA), a biodegradable synthetic polymer, has been widely used in combination with natural carbohydrates and proteins to produce blends with improved mechanical properties (MA et al., 2018; MA; DU; WANG, 2017; ZENG et al., 2019b).

Gelatin, for instance, is a thermally fusible protein also used in film production due to its low cost, biodegradability, and non-toxicity (DJAGNY; WANG; XU, 2001). In this regard, blending gelatin with PVA is an interesting alternative for developing biodegradable films. Moreover, gelatin contains a high amount of proline and hydroxyproline residues, which have an affinity for polyphenols, such as the anthocyanins found in blueberry extract (GARCÍA-ESTÉVEZ et al., 2017; GOMES et al., 2024).

In a previous by study Gomes et al. (2024), PVA-gelatin polymer blends were developed with blueberry extracts and investigated for their capability to change color in response to pH variations. The findings demonstrated that the films exhibited potential as intelligent food packaging. However, further characterization is necessary since, for a polymeric film to be effectively applied as food packaging, it is essential to understand its mechanical and thermal properties, as well as its behavior in the presence of water/humidity, considering that water is a major component of countless food products.

Therefore, the primary objective of the present study is to complement the investigation of PVA/gelatin films with blueberry extracts by characterizing them to determine whether the presence of these extracts, in addition to imparting intelligent properties to the films, enhances their physical-chemical performance.

2. Materials and Methods

2.1 Materials

Blueberry (*Vaccinium myrtillus*) Probst fruits were obtained from the fruit-producing company Quali Fresh (Barbacena, Minas Gerais, Brazil) and stored at -18 ± 2 ºC until analysis. The polymers used in the film preparation were PVA (Sigma Aldrich Co., St. Louis, MO, USA), degree of hydrolysis >99% and molecular weight (Mw) of 85,000 to 124,000 (g/mol), and 99% hydrolyzed, colorless, flavorless gelatin from the Dr. Oetker brand (Lot No. L346036A 22N), food grade.

All chemical reagents used in this study were of analytical grade. Glycerol and ethyl acetate were obtained from Vetec Química Fina Ltda (Recife, PE, Brazil). Ethyl alcohol and methyl alcohol were acquired from Merck (Taufkirchen, Bavaria, Germany). The quantitative paper filter (Unifil, Ø 11) was obtained from Dinâmica (São Paulo, SP, Brazil).

2.3 Methods

2.3.1 Preparation of aqueous concentrated

The extraction of phenolic compounds was carried out according to the methodology described by Gomes et al. (2024). Briefly, the blueberry was crushed in a mixer (RI1602, Philips Walita, Brazil), and mixed with 70% ethanol (v/v), in a 1:10 (w/v) ratio. The mixture was acidified to pH 2.0 with hydrochloric acid (HCl) (37% v/v) and subjected to sonication in an ultrasound bath (Elmasonic TI-H10, Elma, Singen, BW, Germany), at 45 kHz (40 ± 2 °C, 50 min). Subsequently, it was filtered (Whatman No. 1 filter paper) under vacuum and concentrated in a rotary evaporator (RV 10 digital V, IKA, Staufen, BW, Germany), 100 rpm, at 40 ± 2 °C, until the complete solvent evaporation.

2.3.2 Purification of aqueous concentrated blueberry extracts

Purification was done using a C18 separation cartridge (Sep-Pak Vac 35 cc, Waters, Milford, USA) under vacuum according to the methodology described by Gomes et al. (2024). First, the cartridge was conditioned with 50 mL of acidified methanol (0.01% HCl) and 50 mL of acidified distilled water (0.01% HCl). Next, a 50 mL aliquot of the concentrated blueberry extract was loaded into the cartridge. The removal of other compounds (such as sugars) was done with 50 mL of acidified distilled water (0.01% HCl). To obtain the extract rich in phenolic compounds, an additional passage of 50 mL of ethyl acetate was performed, leaving the anthocyanins adsorbed and obtaining the purified phenolic extract. To remove the adsorbed anthocyanins and obtain the purified anthocyanin extract, 50 mL of methanol was loaded into the cartridge (NORATTO et al., 2010; RODRIGUEZ-SAONA; WROLSTAD, 2001b) and the anthocyanins were eluted.

2.2.3. Production of colorimetric indicators (polymer blend based on PVA and gelatin, with blueberry extract)

The blend films (PVA:GL) were produced by mixing two suspensions, 2% (w/v) gelatin in water at 60 ± 2 °C (suspension A) and 2% (w/v) PVA in water at 80 ± 2 °C (suspension B) (GOMES et al., 2024), which was stirred for 5 h, then, mixed and kept under stirring (300 rpm) until cooled to 40 ± 2 °C. The gelatin control film was produced with 4% gelatin and dispersed as suspension A. The PVA control film was produced with 4% PVA and dispersed as suspension B. Glycerol (30% w/w, based on polymer mass) was added in all treatments, and the extracts (10 % (v/v), based on dispersion volume) were added to the blend suspension and homogenized for 20 min, at 25 ± 2 ºC, with magnetic stirring (300 rpm). Subsequently, the suspensions were placed in an ultrasound bath (40 kHz) for 10 min to remove air bubbles, then

poured onto glass plates (33 cm x 9) cm² and dried at 25 ± 5 °C, RH 62 ± 5%, for 24 h and 20 ± 2 °C, RH 53 ± 5% for 72 h in a climatic chamber (420-CLDTS 300, Ethik Technology, Brazil). The films were vacuum packed (200S, Selovac, Brazil) in polyethylene and nylon bags, wrapped in aluminum foil and stored at 20 ± 2 ºC, for subsequent analysis. In total, it was obtained six films: (i) FB film, FPVA:GL incorporated with crude blueberry extract; (ii) FF film, FPVA:GL incorporated with phenolic blueberry extract; (iii) FA film, FPVA:GL incorporated with blueberry anthocyanin extract; (iv) FC film, film produced with glycerol, PVA and gelatin; (v) FG film, film produced with glycerol and gelatin; and (vi) FPVA film, film produced with glycerol and PVA. As described in the study by Gomes et al. (2024) with modifications based on the final color clearly visible with naked eye of the dispersions obtained for each film with blueberry extract.

2.2.4 Experimental design

The experiment was conducted in a completely randomized design. One factor was studied: films produced with different type of blueberry extracts, at 6 levels (FB, FA, FF), and the controls (FC, FG, FPVA).

2.2.5 Microstructural characteristics

The topography of the film surfaces was characterized through SEM images taken in a low vacuum tabletop microscope (Hitachi Hi-Tech, model TM3000, Japan). Samples of each film, with dimensions of (0.2×0.5 cm²), were fixed on stubs with the aid of tweezers on a conductive double-sided carbon tape. The electron-accelerating voltage was used in automatic mode. The magnification of the images obtained was 400×. Then, Energy Dispersive Spectroscopy (EDS) coupled with SEM was performed to determine the chemical elements present on the surface of each film (VIEIRA et al., 2021).

2.2.6 Thermogravimetric analysis (TGA)

TGA was performed using a Thermogravimetric Analyzer (model DTG-60H, Shimadzu, Japan). Approximately 3 mg film pieces were heated from 20 °C to 700 °C, at a rate of 10 °C/min, in a nitrogen atmosphere (50 mL/min). The thermal stability of the samples was descriptively evaluated from thermogravimetric curves analyses.

2.2.7 Grammage

Grammage (G) is defined as the weight (g) obtained from a given area of the material and is usually expressed in g.m-². The films were cut to a size of 0.1x0.1 m² and were subsequently weighed using an analytical balance, thus obtaining the mass (g) of this piece of film. Each repetition of treatment was evaluated in triplicate. and the grammage was calculated with the Equation 1.

$$G\ (g.m^2) = \frac{mass\ (g)}{area\ (m^2)}\ (1)$$

2.2.8 Thickness and mechanical properties

Film thickness was determined using a digital micrometer (Model 547-401, Mitutoyo, Japan, with an accuracy of 0.001 mm). Measurements were taken at ten random film positions, and the mean values were calculated. Tensile strength (TS, in MPa), elongation at break (EB, in mm), and Young's modulus (YM, in MPa) were determined with a Universal Testing Machine (model 3367, Instron Corporation, Norwood, MA, USA) according to ASTM D882 (ASTM, 2012). Film samples (10 x 2.5 cm²) were grabbed by two grips and stretched at a crosshead speed of 500 $mm.min^{-1}$. Each repetition of treatment was evaluated in triplicate.

2.2.9 Moisture (U), Swelling index (I) and Solubility (S)

The initial mass (m0) of the films (1 × 4 cm²) was obtained using an analytical balance (AUY220, Shimadzu, Japan). The film samples were dried at 105 ± 2 °C for 24 h in an air-circulating oven (400-6ND, Ethik Technology, Brazil) and the mass of the dry sample was weighed (m1). The moisture content (U%) of the film was calculated from Equation 2 (TIAN et al., 2021).

$$U(\%) = ((m0 - m1)/m0)) * 100\ (2)$$

Subsequently, the dried film (m1) was immersed in distilled water and incubated at room temperature for 24 h to calculate the extent of swelling of the film sample. Then, excess of water was removed from the surface of the film with the aid of paper towels, and the mass of the sample was weighed on an analytical balance (m2). The swelling index (I%) was calculated from Equation 3 (TIAN et al., 2021).

$$I(\%) = ((m2 - m1)/m1) * 100\ (3)$$

The solubility (S%) of the film was calculated (Equation 4) from the mass of the dried film (105 ± 2 °C, for 24 h) (m1) and the mass of the film immersed in 50 mL of distilled water (25 ± 2 °C, for 24 h) under stirring and dried again (105 ± 2 °C, for 24 h) (m3) (TIAN et al., 2021).

$$S(\%) = ((m1 - m3)/m1)) * 100 \quad (4)$$

2.2.10 Statistical analysis

Data were analyzed using analysis of variance (ANOVA) followed by Scott Knott test, when necessary, (α=0.05) to compare treatments, using R software (R Core Team, 2024) (JELIHOVSCHI; FARIA; ALLAMAN, 2014).

3. Results and Discussion

3.1 Scanning electron microscopy (SEM)

In Figure 1, picture of the films containing blueberry extracts can be observed: FB, film produced with the crude blueberry extract; FF, film produced with the phenolic extract, FA, film produced with the anthocyanin extract, and the controls: FC, film produced with the PVA and gelatin; FG, film produced with the gelatin; and FPVA, film produced with the PVA. Besides, the Figure 2 displays the topography of the films.

Figure 1 - Photography of the films FC, FPVA, FG, FB, FF and FA that were developed and then caracterized.

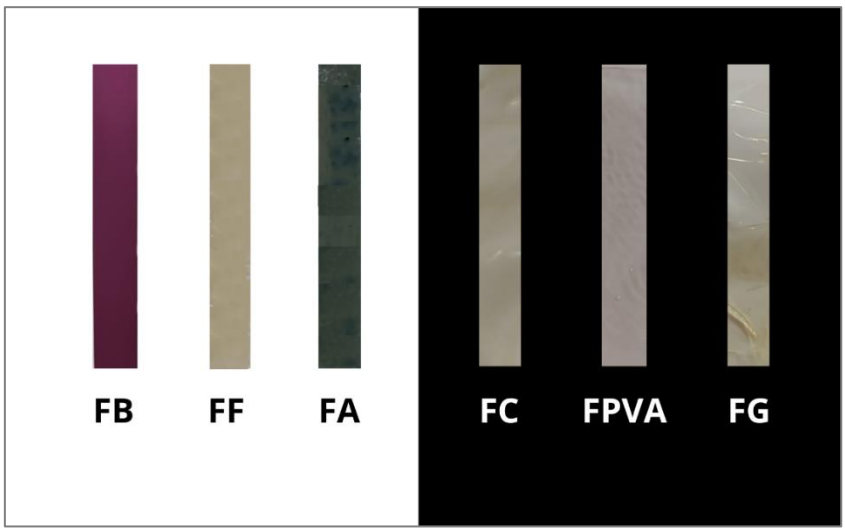
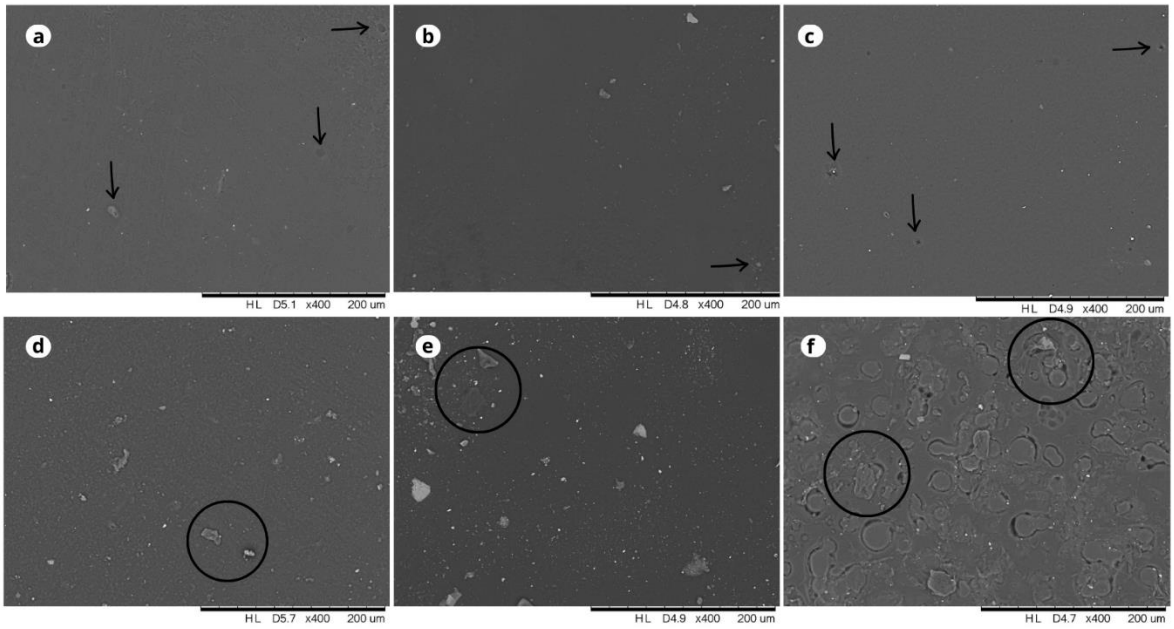


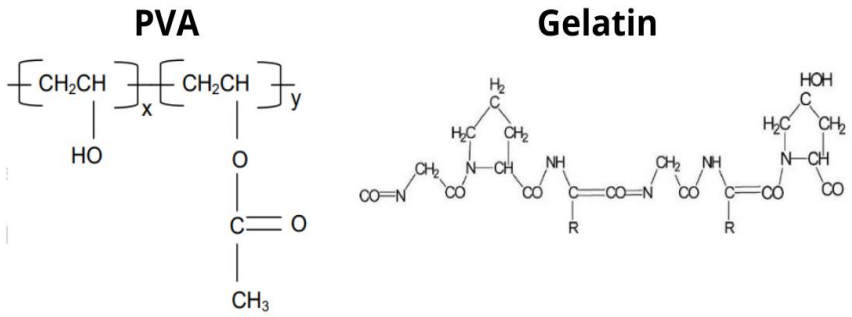
Figure 2 - SEM micrographs of the PVA and gelatin films added with different blueberry extracts. (a) FC; (b) FPVA; (c) FG; (d) FB; (e) FF e (f) FA.



FC (film produced with the PVA and gelatin), FPVA (film produced with the PVA), FG (film produced with the gelatin), FB (film produced with the crude extract), FF (film produced with the phenolic extract), and FA (film produced with the anthocyanin extract). Micropores are indicated by black arrows and spots are highlighted with black circles.

Control films (Figure 2a, b, and c) exhibited a homogeneous, smooth surface with good dispersion, suggesting compatibility between the gelatin and PVA polymers. This can be explained by a strong intermolecular association between the functional groups of GL and PVA. The basic chemical structure of PVA and gelatin is shown in Figure 3.

Figure 3 - The basic structure of PVA and gelatin.



A similar result was obtained in the study of Rashid et al. (2023), in which the surface of the PVA and gelatin film was homogeneous, smooth, non-porous and compact. In the study of

Tymczewska et al. (2022) the gelatin and PVA film showed a relatively uniform fine structure, revealing no signs of phase separation, the pore distribution was nearly uniform, and most pores did not exceed 1.5 µm in diameter. Polymeric blends based on PVA and gelatin are macroscopically homogeneous, colorless and transparent (Figure 1) (KIM; ROY; RHIM, 2022).

The films presented micropores with different shapes (represented by the black arrows in Figure 2), which may be due to the presence of the plasticizer glycerol (30%) in the polymer matrices (TEIXEIRA et al., 2021). The same was found in the films containing PVA, gelatin, glycerol, and mulberry anthocyanin extracts of Zeng et al. (2019), and according to the authors, these micropores may result from small bubbles existing in the polymer dispersion.

The film surfaces were intact, and cracks were not observed in any of the films, which was the same as that observed in the film by Rashid et al. (2023) and was related to the high flexibility of this polymer blend. On the other hand, in the study by Oyeoka et al. (2021), the gelatin and PVA film showed some unbound granules in the micrograph with some discontinuous cracks that may have resulted from inadequate homogenization during film preparation. The authors obtained better interaction and a cohesive structure in the film reinforced with 5 wt % cellulose nanocrystals.

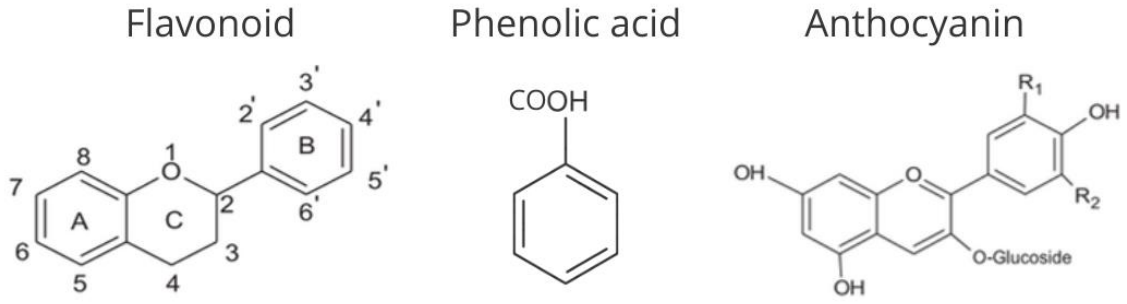
The PVA and gelatin films with blueberry extracts (Figure 2d, 2e, and 2f) exhibited a more heterogeneous surface than the control films (Figure 2a, 2b, and 2c), with some spots presente (highlighted in circles in Figure 2). Some white spots were observed in all films, which may be related to the existence of non-dispersed polymer (LOZANO-NAVARRO et al., 2018). Furthermore, the presence of small agglomerates in films, spots are highlighted with black circles, (Figure 2d and 2e) may be due to incomplete dispersion of the polymers.

It was observed that the surface of the FA, which has the blueberry anthocyanin extract, presented greater heterogeneity than that of the FF and FB films, which have the phenolic extract and the crude blueberry extract, respectively, due to the higher anthocyanin content in the blueberry anthocyanin extract (GOMES et al., 2024). In Figure 2f, the FA exhibited a more heterogeneous surface, this may be due to the lower affinity and interacion of blueberry anthocyanins extracts with the polymer matrix and plasticizer (ZENG et al., 2019b).

The anthocyanins have fewer free hydroxyls than non-anthocyanic phenolic compounds, such as phenolic acids and flavonoids, so they form fewer hydrogen bonds with the PVA-gelatin matrix and glycerol. The basic chemical structures of these phenolic compounds are shown in the Figure 4. Furthermore, this indicated that the anthocyanins present in the polymeric matrix of the FA had predominantly positively charged and the difference in electronegativity with

PVA and gelatin hindered the interaction and favored the formation of a film with a less homogeneous surface.

Figure 4 - The basic structure of flavonoid, phenolic acid and anthocyanin.

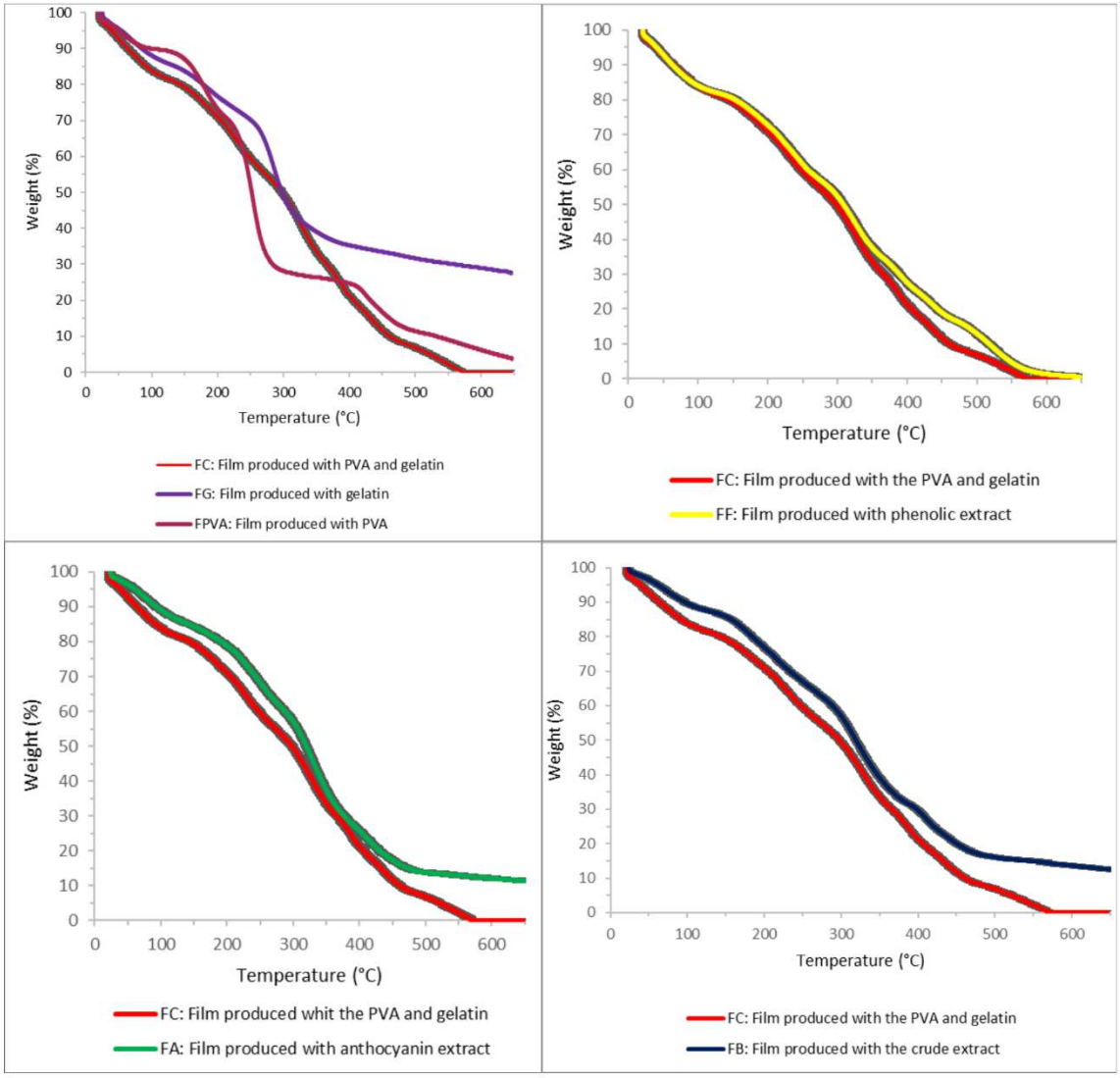


Regarding the EDS Spectrum, the chemical elements present on the surface of the films were, in the majority, carbon, oxygen, and nitrogen, corroborating with the expected based on the chemical structure of PVA, gelatin (Figure 3) and of phenolics compounds phenolics present in blueberry extracts (Figure 4) (OYEOKA et al., 2021; RASHID et al., 2023).

3.2 Thermogravimetric analyses

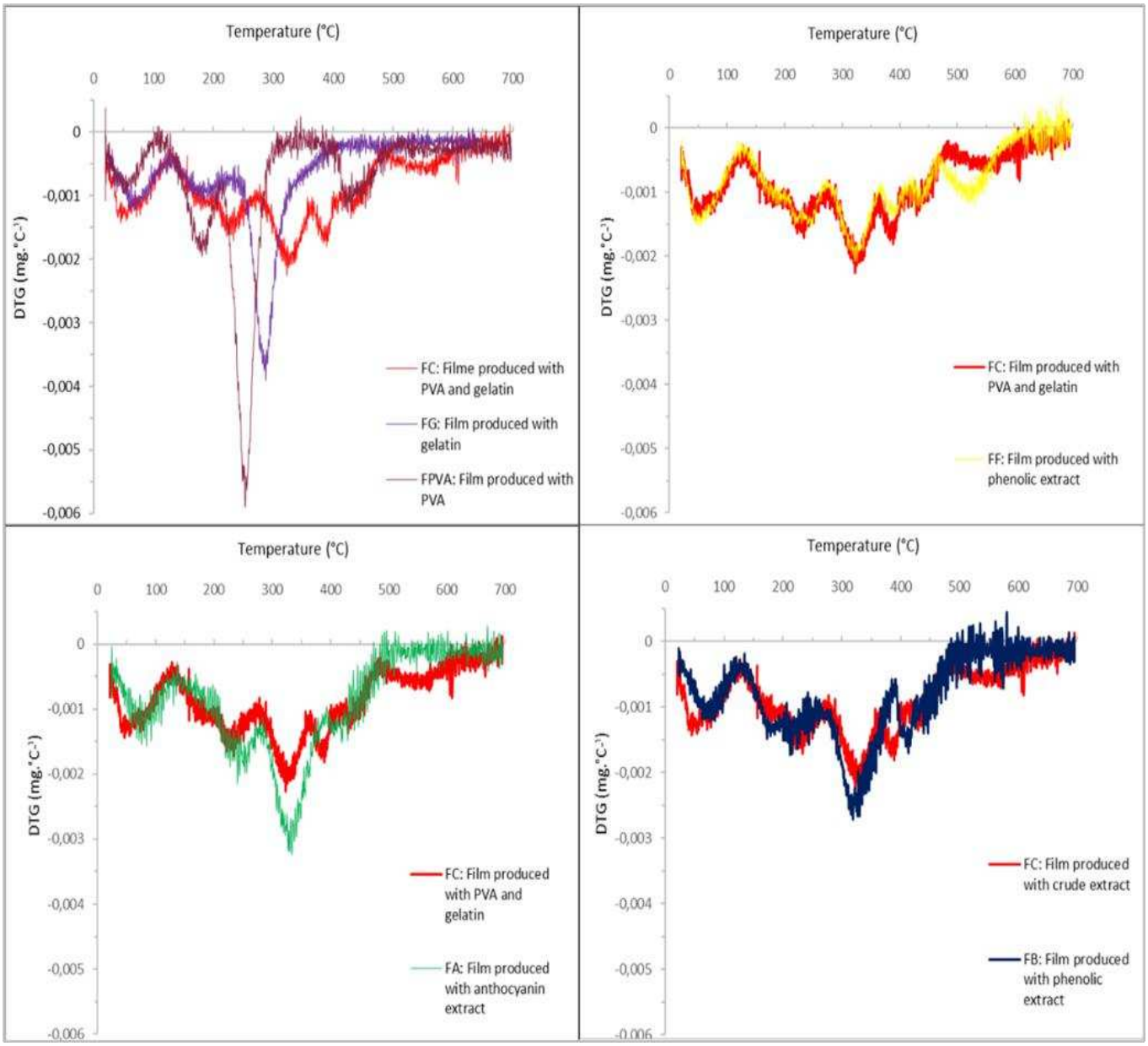
The thermal properties of the films with and without blueberry extracts were investigated based on the thermogravimetric (TG) and derived TG (DTG) curves of the samples, shown in Figure 5 and 6, respectively.

Figure 5 - Thermogravimetric curves (TG) of the films.



FC (film produced with the PVA and gelatin), FPVA (film produced with the PVA), FG (film produced with the gelatin), FB (film produced with the crude extract), FF (film produced with the phenolic extract), and FA (film produced with the anthocyanin extract).

Figure 6 - Thermogravimetric derivative (DTG) (mg/°C) of the films.



FC (film produced with the PVA and gelatin), FPVA (film produced with the PVA), FG (film produced with the gelatin), FB (film produced with the crude extract), FF (film produced with the phenolic extract), and FA (film produced with the anthocyanin extract).

The TG curves expressed in Figures 3 and 4 indicated that the thermal degradation of the films occurred in three stages. For all treatments, the first thermal degradation event occurred between 20 °C and 180 °C, corresponding to approximately 20% mass loss. This stage is mainly related to the evaporation of water and the volatilization of the glycerol plasticizer present in the polymer matrix (FREITAS et al., 2020; QIAO et al., 2024). For films with blueberry

extracts, in the first stage, there is also a loss of low molecular weight and volatile compounds (TEIXEIRA et al., 2022a).

The PVA control film showed a second event at a temperature between 200 °C and 300 °C, with a mass loss of 70% and a maximum degradation temperature of 270 °C. This event can be attributed to the decomposition of PVA (QIAO et al., 2024). The gelatin control film showed a second event at a temperature between 250 °C and 320 °C, with a mass loss of 50% and a maximum degradation temperature of 298 °C, attributed to the decomposition of gelatin (QIAO et al., 2024).

The start temperature of the second thermal event for the blend films (FC) and films with blueberry extracts (FB, FA, and FF) was higher and occurred between 290 °C and 370 °C, with a mass loss of 60%, due to the decomposition of PVA, gelatina, and extracts (BASUMATARY et al., 2018).

The differences in mass variations between the films added with blueberry extracts and the control samples may be due to the loss of mass of residual water present in greater quantities in the films with aqueous extract (FREITAS et al., 2020). These water molecules interact with functional groups of chemical species that may be present in the extracts, such as sugars, acids, phenolic compounds and anthocyanins (FREITAS et al., 2020). The FB, FA and FF have proven to have higher moisture content than FC, FPVA and FG.

The maximum degradation peak increased as the concentration of phenolic compounds in the film increased, suggesting greater thermal stability of the FB and FA. This result may be related the fact that phenolic compounds are effective as a thermal stabilizer due to their radical scavenging activity (OLIVEIRA FILHO et al., 2021; YANG et al., 2022)(YANG et al., 2022). The FB and FA were added to the crude extract and blueberry anthocyanin extract, respectively, both extracts rich in antioxidant phenolic compounds (GOMES et al., 2024).

The third thermal event occurred similarly for all treatments in temperatures above 550 °C. This thermal degradation is due to degradation and carbonization reactions of organic materials (PRIETTO et al., 2017; QIAO et al., 2024; TEIXEIRA et al., 2022b).

In the DTG graph, all decomposition peaks of the polymer blends moved to a slightly higher temperature compared to the films with only one polymer. Thus, it can be concluded that the polymer blend of PVA and gelatin presented the best thermal stability, making its use possibly more advantageous than films with separate polymers in the food packaging area.

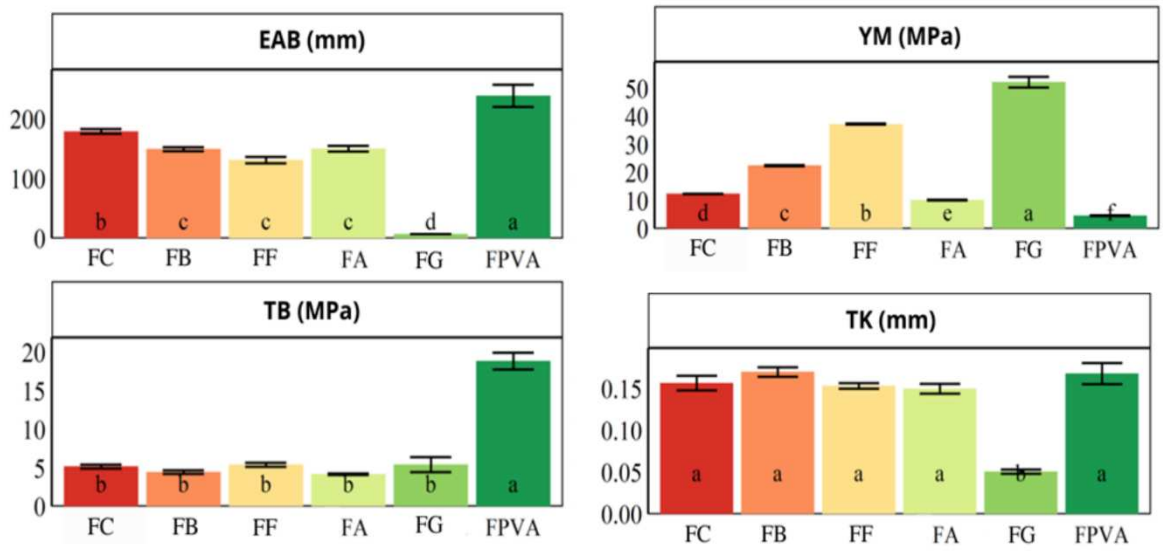
The films with the addition of crude extract and blueberry anthocyanin extract increased the thermal stability of the PVA and gelatin film, and the film with the addition of phenolic extract did not alter the thermal stability of the polymer blend. This indicates that the blueberry

extracts, rich in antioxidant phenolic compounds, positively influenced the thermal properties of the PVA and gelatin polymer blend.

3.3 Thicknesses and mechanical properties

The thickness (TK) and mechanical properties the films were determined and are shown in Figure 7.

Figure 7 - Mechanical properties and thickness of the films.



FC (film produced with the PVA and gelatin), FPVA (film produced with the PVA), FG (film produced with the gelatin), FB (film produced with the crude extract), FF (film produced with the phenolic extract), and FA (film produced with the anthocyanin extract). Means followed by the same letter, in the bars, do not differ statistically using the Scott Knott test at a significance level of 5%. TK: thickness; TB: tensile strength at break; EAB: elongation at break; YM: Young's modulus. All measurements were carried out in triplicate.

The thickness of the investigated films varied between 50 and 170 μm. The TK of the gealtin and PVA films investigated by Haghighi et al. (2021) varied between 42 and 48 μm, being smaller than that of the present study, since the methodology and concentration of the components were different. Haghighi et al. (2021) films were prepared with GL and PVA (3% by weight), in a ratio of 1:1 and glycerol (25 g glycerol/100 g of dry polymer). And different amounts of bacterial cellulose nanowhiskers were also added to the GL-PVA mixture. The films were obtained by casting 20 mL in petri dishes (14.4 cm in diameter).

As shown in Figure 7, the incorporation of blueberry extracts did not promote differences in film thicknesses ($p \geq 0.05$) in relation to the controls FC and FPVA. Furthermore, the gelatin film (FG) had the lowest thickness value (0.05 ± 0.01 mm) ($p < 0.05$) compared to the others. Generally, polymer blends have a lower degree of variability in relation to their thicknesses (RASHID et al., 2023). Similarly, Jebel et al. (2025) found that adding eggplant skin extract did not affect the thickness of gelatin films.

Regarding mechanical properties, the Young Modulus values of the blend film were intermediary, while pristine gelatin films were the most rigid (highest YM; $p < 0.05$), and the PVA film was the least rigid and most flexible (lower YM; $p < 0.05$). In the present study Tymczewska et al. (2022) all the samples were characterized by relatively high flexibility compared to pristine gelatin, related to the presence of PVA acting as the plasticizer in the mixtures.

Concerning the films with extract (FA, FB, and FF), with lower concentrations of anthocyanins, as in FB, the YM increased (GOMES et al., 2024). Phenolic compounds can act as crosslinking agents, strengthening the intermolecular interactions between the polymer chains and, therefore, increasing the material rigidity (JEBEL et al., 2025). Non-anthocyanic phenolic compounds have large number of free hydroxyls and available to interact with the GL-PVA blend film matrix to form hydrogen bonds, resulting in an increase in YM no FB and FF (HAGHIGHI et al., 2021). Being the FF with the highest value due to the predominance of non-anthocyanin phenolic compounds in the phenolic extract incorporated in this film.

The same behavior was observed in the film by Jebel et al. (2025) in gelatin films using eggplant skin extract (ESE). The TS and YM of the gelatin films were significantly increased ($p < 0.05$) with the addition of ESE at lower concentrations (2 - 4 %).

On the other hand, the elongation at break (EAB) and tensile strength at break (TB) of PVA films were the highest ($p < 0.05$), which is desirable for packaging since one of its functions is to protect the packaged product, maintaining its integrity. It was observed that PVA contributed more to the tensile strength of the film than gelatin, as also noted by Oyeoka et al. (2021).

Similarly, blend films, regardless of type blueberry extract addition, presented intermediary values of EAB, while TB values of FC, FB, FA, and FF were statistically equivalent to GL films ($p < 0.05$). In the study by Zeng et al. (2019a), the addition of mulberry anthocyanin extracts (MAE) influenced the TB and EAB of PVA (10% w/v), 1% (v/v) of glycerol and gelatin (2% w/v) films when added at a higher concentration, in which the TB value decreased and EAB increased with the increase in MAE addition from 15 to 45 mg

MAE/100 mL, but at lower concentrations also it did not influence. The decrease in TB and EAB films (FC, FG, FA, FB, and FF) in relation to the FPVA, reduces the material's tenacity, impacting flexibility and resistance under stress (TYMCZEWSKA et al., 2022).

The EAB is an important mechanical property for the application of polymers in packaging. This property is calculated as the percentage increase in length that the film will have achieved before failure (OYEOKA et al., 2021). It is observed that the polymer blend of PVA and gelatin (2% w/v; 1:1) can present higher EAB values (176%) compared to pure gelatin films (4% w/v) (7.91%). A similar result was observed in the study of Dong et al. (2024), when 5% PVA was added to the pure gelatin film, the elongation at break was improved by 76.7 %. In the study carried out by Oyeoka et al. (2021), the authors also observed that PVA contributed more to the increase in the films EAB than gelatin. Our results corroborated, in which the EAB increased in 96% with the addition of PVA.

The addition of blueberry extracts resulted in a significant decrease ($p<0.05$) in EAB of FB, FF and FA films, compared to FC. The same occurred in the study of Tymczewska et al. (2022) where addition of the black cumin cake extract to the G-PVA blend (5% w/v, 5:3 v/v) resulted in a slight decrease in the EAB (125.16%) in comparison with the control film (EAB = 137.03%). In polymer films, elongation capacity is generally increased by the presence of plasticizers (such as glycerol). However, blueberry extracts can compete with the plasticizer for interactions with the matrix, reducing its effect. Consequently, the material became more susceptible to destruction while stretching (TYMCZEWSKA et al., 2022).

The TB and YM of the FC, with values of 5.22 and 12.55 MPa, respectively. Rashid et al. (2023) developed a PVA (1.25%), gelatin (3.75%) and glycerol (25% w/w based on the mass of polymers) film with higher TB (30.20 MPa) and lower YM (0.59 MPa) values than the present study. Thus, Rashid et al. (2023)obtained a polymer blend with greater elasticity and more mechanical resistance, these characteristics being interesting for the applicability tested by them in dressings. The characteristics of this film were closer to the characteristics obtained for the FPVA film of this study.

Considering the application of films as colorimetric seals indicating pH changes (GOMES et al., 2024), this reduction in film properties is less relevant. Furthermore, although greater tensile strength is desirable, it is also important to note that the strength of packaging materials needs to be considered so that sustainable and biodegradable packaging has a more limited shelf life in order to minimize environmental pollution (OYEOKA et al., 2021).

High concentrations of anthocyanins in films, as in the case of FA (GOMES et al., 2024), reducing the rigidity of the film. This occurs because anthocyanins and the presence of

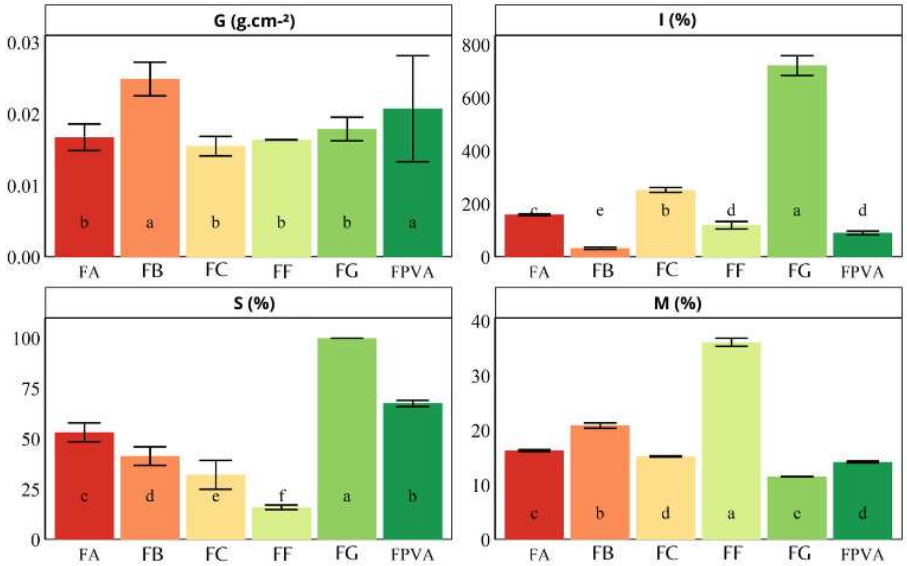
predominantly positively charged in their molecule interfere with the interactions between the polymer chains of PVA and gelatin, occurring electrostatic repulsion with gelatin (positively charged), reducing the intermoleculars forces and increasing the movement of the molecules and, consequently, reducing the Young modulus (reducing the coesion the material) but it was not enough to increasing its elasticity (EAB) (JEBEL et al., 2025; TYMCZEWSKA et al., 2022). The same behavior was observed in the film by Jebel et al. (2025) in gelatin films using eggplant skin extract (ESE), rich in anthocyanins. The incorporating of 8 % ESE resulted in a decrease in YM values.

In summary, the YM of gelatin and PVA - based films are influenced by the type of the added blueberry extract. The FB, FA, and FF films were favorable for the development of sustainable active and intelligent films, as they maintained similarly the mechanical properties (TB and TK) of the PVA and gelatin blend, which were improved in relation to FG by presenting greater flexibility.

3.4 Physical and Chemical properties

The moisture content (M), swelling index (I), solubility (S), and grammage (G) of control films and PVA and gelatin films added with blueberry extract are displayed in Figure 8.

Figure 8. Moisture content (M), solubility (S), grammage (G) and swelling index (I) of control films and films added with blueberry extract.



FC (film produced with the PVA and gelatin), FPVA (film produced with the PVA), FG (film produced with the gelatin), FB (film produced with the crude extract), FF (film produced with the phenolic extract), and FA (film produced with the anthocyanin extract). Means followed by

the same letter, in the bar, do not differ statistically according to test Scott Knot at a significance level of 5%.

According to the data presented, the M values of the prepared films ranged from 11.55% to 36.17%. Phenolic acids and flavonoids generally have more free hydroxyl groups (-OH) than anthocyanins, these hydroxyls form more H-bonds with water the polymer suspension, keeping the film moist. Thus, the FF that was added with blueberry extract rich in phenolic compounds (except anthocyanins) has a higher moisture content and the FA that was added with blueberry extract rich in anthocyanins has the lowest moisture content. The FB was incorporated with the crude extract, rich in phenolic acids, flavonoids and anthocyanins, and the interaction occurs more between these compounds than with water, which is why the lower moisture content in relation to the FF is higher than that of the FA. In the study of Tymczewska et al. (2022), the moisture content of the FC (5% w/v PVA and 5% w/v gelatin; 5:3 v/v) was 18.09%, value close to that found in this study of 15,4%.

Gelatin and PVA are hydrophilic polymers, meaning that films formed by these polymers have an affinity for water. PVA and gelatin hydrophilicity is due to several hydroxyl groups that make it highly polar and, therefore sensitive to water molecules (OYEOKA et al., 2021). A relatively high moisture value in pure gelatin film can be explained by the presence of a large number of OH groups. In this way, the water entry into the polymer matrix caused the PVA and gelatin films to swell (RASHID et al., 2023). Furthermore, the film formed with pure gelatin had the highest I (%) due to the repulsion between adjacent amines in gelatin, which become positively charged in water and may have increased the swelling rate (PAL; BANTHIA; MAJUMDAR, 2007).

The FB had a higher grammage and this may have influenced its low swelling index due to the slower penetration of water into the film, and also because it probably has fewer empty spaces for water to enter and form hydrogen bonds. The phenolic compounds present in the blueberry extracts decreased the water swelling index of the control polymer blend FC (ZHAI et al., 2017), as these compounds bind to the free hydroxyl groups of the matrix and the glycerol, restricting the mobility of the polymer matrix and making it difficult for water to penetrate the polymer matrix (BELBEKHOUCHE et al., 2011). In addition, FA presented the highest I (%) among the films with blueberry extract due to the electrostatic repulsion between the anthocyanins present predominantly with positive charges and the adjacent amines in gelatin, which become positively charged in water and may have increased the swelling rate.

The film with FF presented the lowest solubility in water, this can be attributed to the strong interaction of non-anthocyanic phenolic compounds with the GL-PVA chains and with

glycerol, through hydrogen bonding, leading to a lower availability of free hydroxyl groups and, consequently, a reduction in the film's ability to interact with water, reducing solubility (JEBEL et al., 2025). On the other hand, FB and FA presented greater solubility than FC because, in addition to the anthocyanins present in the films having a lower affinity with the polymer matrix and glycerol and making fewer H-bonds, they presented a predominantly positive charge similar to gelatin, which hinders this interaction due to electrostatic repulsion. This space in the polymer matrix favors the entry and interaction of water in the film, increasing solubility. Since FA has more anthocyanins than FB, the solubility of this film was greater.

The physical and chemical properties of gelatin and PVA - based films are influenced by the type of the added blueberry extract. The combination of PVA and gelatin formed a film with less dissolution compared to FG and FPVA, proving to be favorable for application as food packaging. The reduced water solubility of polymer blends is a valuable characteristic for maintaining packaging integrity after exposure to liquid exudate from packaged foods, with emphasis on the gelatin and PVA - based films added blueberry phenolic extract. It is desirable that indicator films be resistant to moisture, especially for use in smart food packaging, due to the large amount of water present in many foods.

4. Conclusion

The FB, FA and FF films appear to be favorable for the development of active and smart films that are sustainable and thermally stable, due to the antioxidant capacity of the extracts rich in phenolic compounds added to the PVA and gelatin matrix.

After the addition of blueberry extracts, the films (FB, FA and FF) maintained the mechanical properties (TK and TB) of the PVA and gelatin blend, which were improved in relation to FG due to their greater flexibility.

The I of the films decreased after the addition of blueberry extracts which was positive for the future application of these films in foods, such as fish and meat, which have a high percentage of water in their composition and undergo pH changes with degradation.

The non-anthocyanic phenolic compounds, phenolic acids and flavonoids, present in the blueberry phenolic extract added to the FF and in the crude blueberry extract added to the FB, have free hydroxyls available to interact with the matrix of the GL-PVA-glycerol blend film to form hydrogen bonds. Thus, FF and FB presented higher moisture content, lower swelling index, lower solubility and greater rigidity than FA (added with blueberry extract with anthocyanins). The FB stood out because, in addition to presenting good physical, chemical and

mechanical properties, it presented better thermal stability than FF, due to the higher content of antioxidant phenolic compounds in the crude blueberry extract.

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Declaration of Conflict of Interest

There are no conflicts of interest to declare.

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Figure captions

Figure 1 - Photography of the films FC (control film), FPVA (PVA film), FG (gelatin film), FB (film produced with the crude extract), FF (film produced with the phenolic extract), and FA (film produced with the anthocyanin extract).

Figure 2 - SEM micrographs of the PVA/gelatin films added with different blueberry extracts. (a) FC; (b) FPVA; (c) FG; (d) FB; (e) FF e (f) FA.

Figure 3 - The basic structure of PVA and gelatin.

Figure 4 - The basic structure of flavonoid, phenolic acid and anthocyanin.

Figure 5 - Thermogravimetric curves (TG) of the films.

Figure 6 - Thermogravimetric derivative (DTG) (mg/°C) of the films.

Figure 7 - Mechanical properties and thickness of the films.

Figure 8 - Grammage (G), swelling index (I), solubility (S), and moisture content (U) of controls and PVA and gelatin films added with blueberry extract.

5. ARTICLE 3: Analysis of the stability and color reversibility of gelatin/polyvinyl alcohol-based colorimetric indicators incorporating blueberry extracts for monitoring the freshness of tilapia fillet and shrimp

Abstract

Two colorimetric indicators based on poly(vinyl)alcohol (PVA) and gelatin (GL) were developed: one incorporated with crude blueberry extract (FB) and the other with purified blueberry anthocyanin extract (FA). Innitialy, they were produced using the casting method and, after drying, were exposed to acidic (acetic acid) and basic (ammonia) vapors. The color variations were quantified using a Hunter LAB colorimeter. The chromatic transition of the FB indicators occurred when exposed to ammonia vapors (30 min), while the FA indicators changed under acid vapors (30 min), both showing visually perceptible alterations. The reversibility of the color transition of the films was also studied by exposure to ammonia vapors (30 min), and by exposure to acetic acid (30 min), as well as the colorimetric stability during storage at 25 ± 2 ºC, which was verified in the presence and absence of light and oxygen. The films were also used as indicator labels fixed to the surface of a lid to separately monitor the quality of fresh shrimp and tilapia fillets, placed in a polypropylene container, and monitored for 7 days under refrigeration (6.5 ± 2 °C). The pH values were determined in samples rightly after the acquisition and after 7 days. The FA indicators changed from blue to purple in the presence of acid vapors and from green to brown in response to the degradation of shrimp and fish fillets, with color difference values (ΔE) of 10.05, 6.38, and 6.01, respectively. FB changed color from purple to green and from purple to brown, and this colorimetric transition was more noticeable than that of FA indicators (higher ΔE), indicating deterioration of shrimp and tilapia fillet, respectively, after 7 days of refrigerated storage. For shrimp, FA and FB indicators changed color concomitantly with the pH value (7.93) considered inappropriate for consumption. For fish fillet, FA and FB indicators changed color with pH (6.83) after 7 days of storage (6.5 °C). Both colorimetric indicators developed were effective for monitoring the freshness of shrimp and tilapia fillets, being a sustainable alternative for monitoring the quality of these foods and reducing waste. The FB indicator stands out for being more stable during storage, sensitive, and with the most intense irreversible colorimetric change, easy to visualize to the human eye when applied to monitoring fresh tilapia fillets and gray shrimp.

Keywords: Blueberry; Fish; Smart film; PVA; Gelatin; Spoilage.

1. Introduction

Colorimetric indicators are substances that can be incorporated into polymeric materials, for the production of intelligent packaging that allows direct communication with the consumer and informs about the current state of conservation of the packaged food, through the visible change in its colors (EZATI; RHIM, 2020a, 2020c; KOSHY et al., 2021; ZENG et al., 2019b). These materials are generally impregnated with dyes that are sensitive to changes in the food environment or the internal atmosphere of the packaging (ZENG et al., 2019b). Currently, several studies report on the development of colorimetric indicators for application in food, which uses plant extracts rich in natural pigments, as they are considered safer for consumers (AYDOGDU EMIR, 2023; LI et al., 2023; LUO et al., 2023; WANG et al., 2023).

Plant extracts can be rich in anthocyanins, a natural pigment that is sensitive to changes in the pH of the medium, causing structural changes that affect the wavelength of light reflected by this compound (JAYAKUMAR et al., 2019; KOSHY et al., 2021). Among the existing sources, blueberries stand out for containing high concentrations of anthocyanins, the main ones found in this fruit being cyanidin-3-O-glucoside, delphinidin-3-O-galactoside, delphinidin-3-O-glucoside, delphinidin-3-O-arabinoside, cyanidin-3-O-galactoside, cyanidin-3-O-arabinoside, petunidin-3-O-galactoside, petunidin-3-O-glucoside, petunidin-3-O-arabinoside, peonidin-3-O-galactoside, peonidin-3-O-glucoside, peonidin-3-O-arabinoside, malvidin-3-O-galactoside, malvidin-3-O-glucoside, malvidin-3-O-arabinoside, and malvidin-3-O-xyloside (JARA et al., 2019; PAES et al., 2014; PIRES et al., 2020). Thus, both raw and purified blueberry extracts can be used to produce colorimetric indicators.

Recently, several biopolymers have been used as polymeric matrices for the development of colorimetric indicators. Among them, polyvinyl alcohol (PVA) has been widely used together herewith proteins, such as gelatin, to improve water sensitivity, reduce costs, and produce films with distinct mechanical properties (DJAGNY; WANG; XU, 2001; MA et al., 2018; MA; DU; WANG, 2017; ZENG et al., 2019b). Gelatin is low-cost and biodegradable, and it also contains a large amount of proline and hydroxyproline residues, which have an affinity with the anthocyanins present in blueberries, facilitating the incorporation of blueberry extract into gelatin films (GARCÍA-ESTÉVEZ et al., 2017). Thus, this work aimed to develop smart films based on gelatin, PVA, and blueberry extracts, raw or purified, to evaluate the sensitivity and colorimetric stability of these potential colorimetric indicators and analyze their effectiveness when applied to tilapia fillet and fresh shrimp.

2. Material and methods

2.1 Material

Blueberry (*Vaccinium myrtillus*) Probst fruits were obtained from the fruit-producing company Quali Fresh, Barbacena, Minas Gerais, Brazil and stored at -18 ± 2 ºC until analysis. The polymers used in the film preparation were PVA (Sigma Aldrich Co., St.Louis, MO, USA), degree of hydrolysis of +99% and molecular weight (Mw) of 85.000 to 124.000 (g/mol), and 99% hydrolyzed, colorless, flavorless gelatin from the Dr.Oetker brand (Lot No. L346036A 22N), food grade, obtained from local store (Viçosa, MG, Brazil). Vacuum-packed headless gray shrimp (class A) and tilapia fillets, both fresh and kept under refrigeration, were purchased from a supermarket in the city of Viçosa, Minas Gerais, Brazil.

All chemical reagents used in this study were of analytical grade. The following reagents were purchased from Sigma Aldrich Co. (St.Louis, MO, USA): ammonia, methanol, and ethanol. Glycerol and acetic acid were obtained from Vetec Química Fina Ltda (Recife, PE, Brazil). The quantitative paper filter (Unifil, Ø 11) was obtained from Dinâmica (São Paulo, SP, Brazil).

2.2. Methods

2.2.1 Preparation of aqueous concentrated blueberry crude extract

The extraction of phenolic compounds was carried out according to the methodology described by Rocha et al. (2018), with modifications. Briefly, the blueberry was crushed in a mixer (RI1602, Philips Walita, Brazil) with 70% ethanol (v/v) in a 1:10 (w/v) ratio. The mixture was acidified to pH 2.0 with hydrochloric acid (HCl) (37% v/v) and subjected to sonication in an ultrasound bath (Elmasonic TI-H10, Elma, Singen, BW, Germany) at 45 kHz (40 ± 2 °C, 50 min). Subsequently, it was filtered (Whatman No. 1 filter paper) under vacuum and concentrated in a rotary evaporator (RV 10 digital V, IKA, Staufen, BW, Germany), 100 rpm, at 40 ± 2 °C, until the complete ethanol evaporation.

2.2.2 Purification of aqueous concentrated blueberry extract

Purification was done using a C18 separation cartridge (Sep-Pak Vac 35 cc, Waters, Milford, USA), under vacuum (25 °C). First, the cartridge was conditioned with 50 mL of acidified methanol (0.01% HCl) and 50 mL of acidified distilled water (0.01% HCl). Next, a 50 mL aliquot of the concentrated blueberry crude extract was loaded into the cartridge. The removal of other compounds (such as sugars) was achieved using 50 mL of acidified distilled water (0.01% HCl). To remove other phenolic compounds, an additional passage of 50 mL of

ethyl acetate was performed, leaving the anthocyanins adsorbed. To remove the adsorbed anthocyanins and obtain the purified anthocyanin extract, 50 mL of methanol was loaded into the cartridge (NORATTO et al., 2010; RODRIGUEZ-SAONA; WROLSTAD, 2001b), and the anthocyanins were eluted.

2.2.3 Production of colorimetric indicators (polymer blend based on PVA and gelatin, with blueberry extract)

The films were produced by mixing two suspensions: 2% gelatin (suspension A) and 2% PVA (suspension B) (Maria et al., 2008; Gomes et al., 2014). Suspension A was dispersed in water at 60 ± 2 °C, and suspension B was dispersed in water at 80 ± 2 °C, for 5 h. Then, the suspensions were mixed and kept under stirring (300 rpm) until cooled to 40 ± 2 °C. Glycerol (30% w/w, based on polymer mass) and extracts were added to the suspension, with the obtention of 2 films: (i) FB film, incorporated with blueberry crude extract (2.5% (v/v)), (ii) FA film, incorporated with blueberry anthocyanin extract (7.5% (v/v)), as described in the study by Gomes et al. (2024) with modifications based on the final color of the dispersions obtained for each film. The control film (without extract addition) was prepared with water and glycerol. The suspensions were homogenized for 20 min, at 25 ± 2 ºC, with magnetic stirring (300 rpm), to produce film-forming suspensions. Subsequently, they were placed in an ultrasound bath (40 kHz) for 15 min to remove air bubbles and poured onto glass plates (33 cm x 9) cm² to dry (25 ± 5 °C, RH 62 ± 5%, for 24 h, and 20 ± 2 °C, RH 53 ± 5% for 72 h in a climatic chamber) (420-CLDTS 300, Ethik Technology, Brazil). The films were vacuum packed (200S, Selovac, Brazil) in polyethylene and nylon bags, wrapped in aluminum foil and stored at 20 ± 2 ºC, for subsequent analysis (GOMES et al., 2024).

2.2.4 Experimental design

The experiment was conducted in a completely randomized design. To evaluate the colorimetric stability, one factor was studied for each type of film (FA and FB), the storage conditions, at 5 levels (shortly after manufactured (T0), in the light and oxygen presence (LO), in the light and oxygen absence (A), in the light presence and oxygen absence (L) and in the light absence and oxygen presence (O). To evaluate the colorimetric sensitivity, and reversibility of colorimetric indicators under a vapor atmosphere (acidic or basic) one factor was studied for each type of film (FA and FB), the storage conditions, at 3 levels (shortly after under volatile ammonia contact, shortly after under volatile acetic acid contact and without contact with any volatile compound (Ambient). To evaluate the colorimetric sensitivity of

colorimetric indicators in shrimp, one factor was studied for each type of film (FA and FB), and for the shrimp, the time storage, at 2 levels (T0 (before storage - on the day of collection of the food at the local market) and T7 (after 7 days of storage at 6.5 °C)). The colorimetric sensitivity of colorimetric indicators in Tilapia fillet was evaluated in the same way.

2.2.5 Color stability of colorimetric indicators

The colorimetric indicators were evaluated regarding time-dependent color stability, lighting, and oxygen. Samples (3 x 3 cm²) were stored at 25 ± 3 °C and 55 ± 5 % RH in the presence or absence of natural light and oxygen. The color stability of FA was observed for 60 days, and 305 days for FB. Then, the C*, h°, and ΔE* were calculated according to Equations (1) – (3), respectively.

The color of the indicator films was characterized using a colorimeter (Color Quest XE, Hunter lab, USA), under the following conditions: D65 illuminant, 10º observation angle, and CIELAB system (L*, a*, b*) and CIELCh system (L*, C*, hº). The colorimetric coordinates [L* luminosity (0 = black and 100 = white), a* intensity from green (-) to red (+), and b* intensity from blue (-) to yellow (+)] were determined. Using the values of the colorimetric coordinates, the chromatic tone angle (h°, 0°/360°=Red, 90°=yellow, 180°=green, and 270°=blue) was calculated according to Equation 1, the Chroma (C*, color intensity or saturation that varies from gray to saturated color) according to Equation 2 (WROLSTAD; DURST; LEE, 2005), and the global color difference (ΔE) according to Equation 3 (POURJAVAHER et al., 2017).

$$h° = arctan(b*/a*) \quad (1)$$

$$C* = \sqrt{a*^2 + b*^2} \quad (2)$$

$$\Delta E = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2} \quad (3)$$

In which ΔL represents the variation of the coordinate L* ($\Delta L* = L* - L0*$), Δa represents the variation of the coordinate a* ($\Delta a = a* - a0*$), e Δb represents the variation of the coordinate b* ($\Delta b = b* - b0*$). L*, a* e b* represents the colorimetric coordinates of the indicator films after storage and L0*, a0* e b0* represents the colorimetric coordinates of the indicator films before storage.

2.2.6 Color sensitivity and reversibility of colorimetric indicators

The sensitivity of FB was evaluated amid exposure of the film to basic vapors and of FA amid exposure to acidic vapors. According to a previous study by Gomes et al. (2024), the PVA and gelatin film with blueberry anthocyanin extract presented a complete color change over time under acidic vapors. The PVA and gelatin film with blueberry crude extract exhibited significant differences in color parameters over time under basic vapors (GOMES et al., 2024).

The FB indicator was placed in a desiccator containing a beaker with 5 µL of ammonia, and its color transition was monitored and measured after 30 min (L*, a*, b*, h°, C*, and ΔE). Then the FB was placed in a desiccator containing a beaker with 5 µL of acetic acid for 30 min, and the color transition was monitored again.

The FA indicator was placed in a desiccator containing a beaker containing 5 µL of acetic acid, and its color transition was monitored and measured after 30 minutes (L*, a*, b*, h°, C*, and ΔE). Then, the FA was placed in a desiccator containing a beaker with 5 µL of ammonia for 30 min and the color transition was monitored again. All measurements were performed in triplicate

The detection limit (LOD) was considered the shortest exposure time of the films to acidic and basic vapors which the overall color difference was above 3.5 ($\Delta E > 3.5$), a value considered visually noticeable by experienced and inexperienced observers (FERNÁNDEZ-RAMOS et al., 2023; MOKRZYCKI; TATOL, 2011).

2.2.7 Application and evaluation of the colorimetric sensitivity of indicators in shrimp and tilapia fillet

To evaluate the colorimetric sensitivity of the indicator films to changes in food freshness over 7 days of storage, 70 g of each food was placed separately in a plastic container (polypropylene) (ERFIZA et al., 2021). A sample of (2.0 x 1.0) cm^2 of the indicator films (FA and FB) was attached to the inner surface of the package lid with the aid of adhesive tape, leaving a large part of the film in direct contact with the atmosphere inside the package (JEBEL et al., 2025) (Figure 1).

Figure 1 - Photograph of the system developed for tilapia fillets and shrimp.

Then, the containers with the shrimp and fish fillets were individually wrapped in PVC film and sealed in polyethylene nylon. The systems were stored at refrigeration temperatures (6.5 ºC ± 2.0 ºC / RH 66 ± 2.0 %), in a glass-lid display refrigerator, which was exposed to artificial light for seven days. The colorimetric change of the films (FA and FB) was determined by colorimetry before (L0*, a0*, b0*, h°, C*, and ΔE) and after 7 days of storage (L*, a*, b*, hº, C*, and ΔE), in three replicates (JEBEL et al., 2025; MA; DU; WANG, 2017).

2.2.8 pH

The pH of the tilapia fillets and shrimps was determined according to the 017/IV method of the Adolfo Lutz Institute (2008). To carry out the analysis, 10 g of sample was crushed and homogenized in 100 mL of distilled water (TEIXEIRA et al., 2022b). The pH of the tilapia fillets and shrimps was measured directly before and after of 7 days storage, using a pHmeter (PG 1800, Gehaka, Brazil), previously calibrated with a pH 7.0 buffer and 10.0, at room temperature (25 ± 2 ºC).

2.2.9 Monitoring shrimp and Tilapia fillets degradation

The monitoring of the degradation of fresh shrimp and Tilapia fillets during storage was performed by determining the pH (item 2.2.8) and the sensory characteristics of odor and color (item 2.2.5). The analyses were performed in 3 replicates, in two times: T0, on the day of collection of the food at the local market and T7, after 7 days of storage.

2.2.10 Statistical Analysis

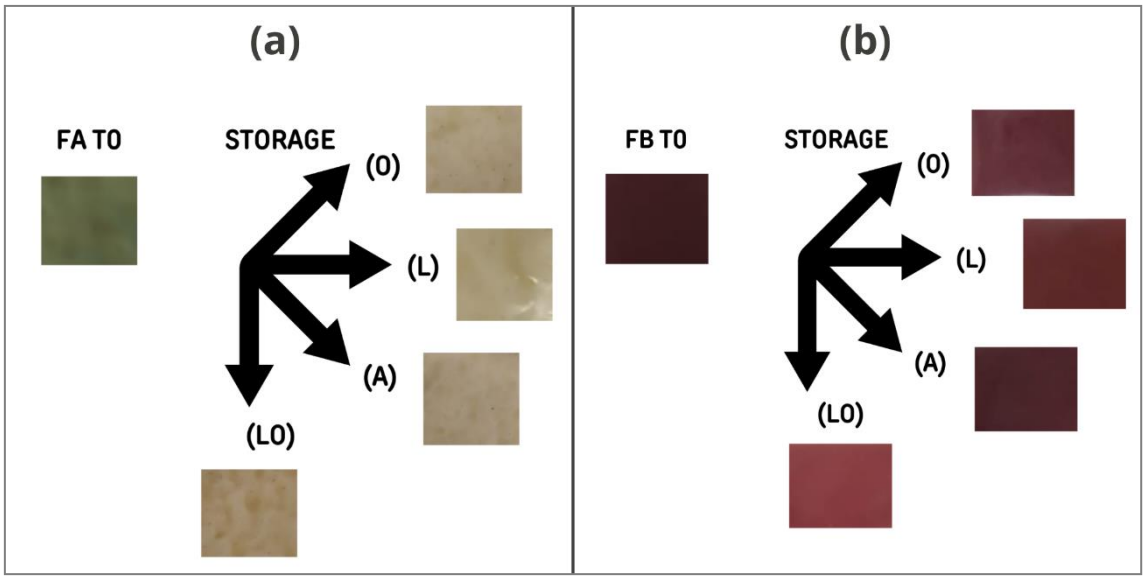
Data were analyzed using analysis of variance (ANOVA) followed by Scott Knott test and t Student test ($\alpha = 0.05$) to compare treatments using R software (R Core Team, 2024). All results were expressed as mean ± standard deviation.

3. Results and Discussion

3.1 Color stability of colorimetric indicators

The color stability of colorimetric indicator films is directly related to the stability of anthocyanins present in the polymer matrix (FREITAS et al., 2020). Images of the FA and FB indicators can be seen in Figure 2, which shows the color of the film immediately after manufacture and after 60 or 305 days of storage, in the presence and/or absence of light and/or oxygen.

Figure 2 - a) FA b) FB photographs shortly after manufactured (T0), after 60 days (FA) or 305 days (FB) the storage in the light and oxygen presence (LO), after 60 days (FA) or 305 days (FB) the storage in the light and oxygen absence (A), after 60 days (FA) or 305 days (FB) the storage in the light presence and oxygen absence (L) and after 60 days (FA) or 305 days (FB) the storage in the light absence and oxygen presence (O).



The variation of the color coordinates L*, a* and b*, and of the color attributes C*, h°, and ΔE* of the FA and FB indicators, stored in the presence or absence of light and oxygen, for 60 and 305 days, respectively, is represented in Figure 3 and 4.

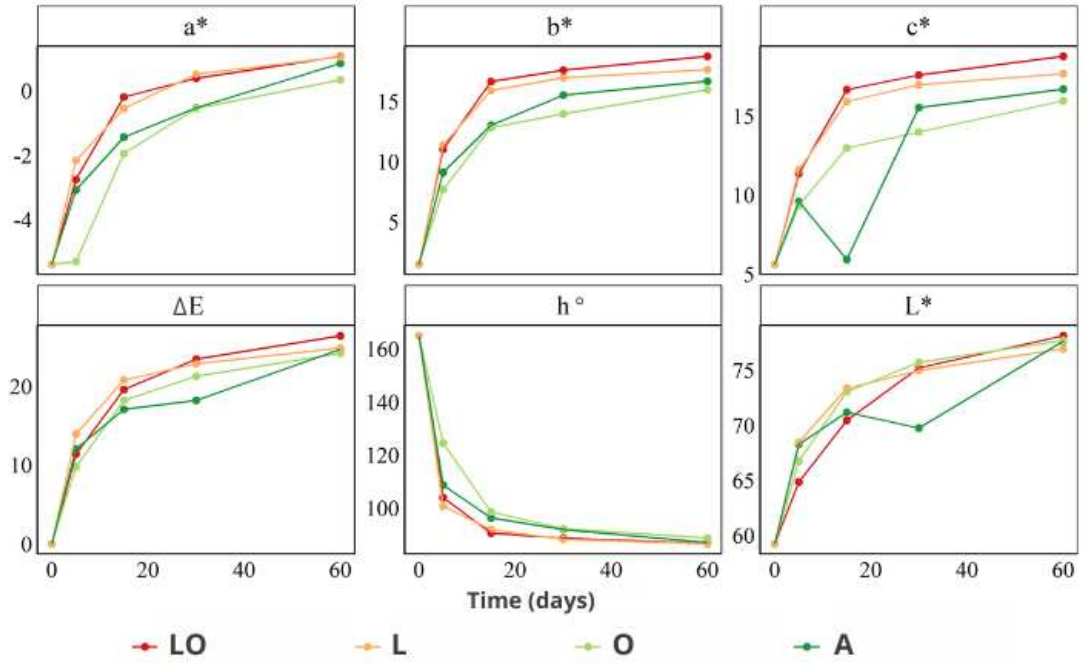
According to Schubring (2002), ΔE values greater than 3 are classified as “very pronounced differences for the human eye”. All conditions to which the FA and FB indicators were exposed resulted in changes in material color, whose ΔE values were greater than 3 after five days of storage, indicating clear and visible changes in the film color. More significant changes were observed for the FA indicators (Figure 2), indicating higher sensitivity of this

film to storage conditions. This behavior can be explained by the fact that the FA indicator was produced with purified blueberry extract. In other words, it consists mainly or entirely of anthocyanin, with sugars and other phenolic compounds removed during the purification process using a C18 column. On the other hand, the FB indicator, produced by incorporating crude blueberry extract, underwent no purification and maintained all these compounds in its composition, which enhanced anthocyanin stability (GOMES et al., 2024).

Anthocyanins are phenolic compounds that are very sensitive to storage time and the presence of light and oxygen. In the absence of other compounds (natural copigments) in the purified extract, anthocyanins become even more sensitive and exposed in the polymeric matrix (CORTEZ et al., 2017; LOPES et al., 2007; SENDRI et al., 2023; XUE et al., 2024). This sensitivity can be explained by its chemical structure, which contains a highly reactive and unstable flavylium ring under different environmental conditions. Anthocyanins remain susceptible to oxygen molecules due to their unsaturated chemical structure (VERMA; SHARMA; MALHOTRA, 2023). In the presence of oxygen, oxidation occurs by the direct mechanism or by the action of oxidizing enzymes, leading to degradation and loss of color (ENARU et al., 2021). In the presence of light, photochemical reactions are induced and break down the flavonoid structure (ENARU et al., 2021). Although antioxidant compounds help stabilize anthocyanins by acting as free radical scavengers, once purified, anthocyanins lose this protection and become more vulnerable to chemical degradation (OANCEA, 2021). Furthermore, anthocyanins are mainly responsible for the coloration of PVA and gelatin films; thus, their stability directly affects the film's color retention (FREITAS et al., 2020).

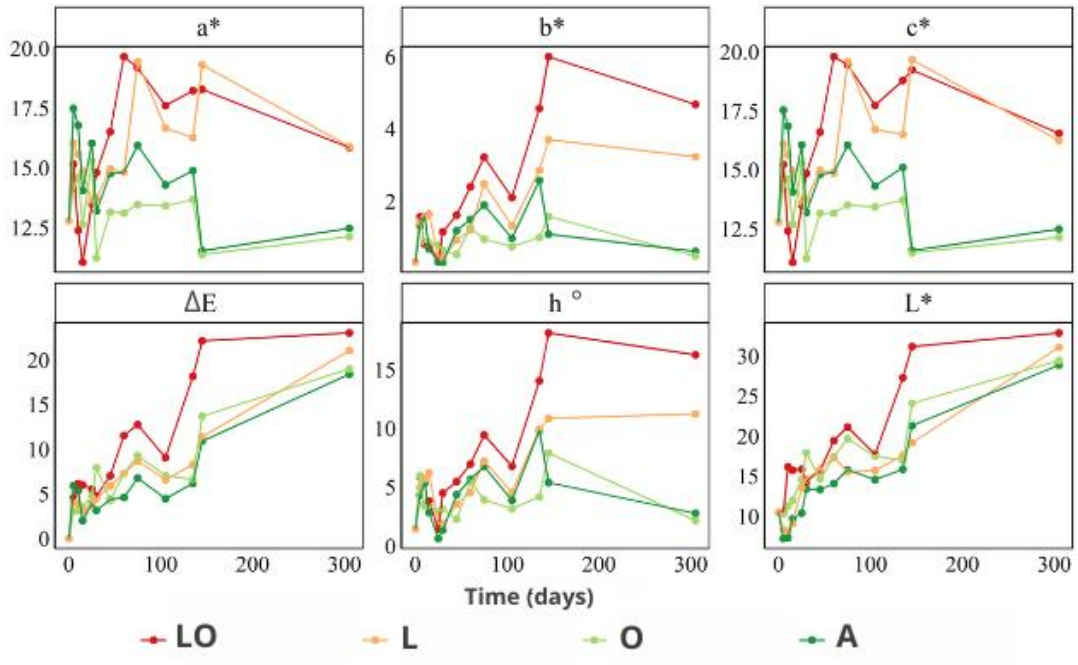
The values of the color coordinate a* and b* of the FA indicator increased while the value of the h° attribute decreased throughout storage (60 days). In other words, the FA indicator changed its initial color from green to yellow over time, as illustrated in Figure 2-a and Figure 3, indicating degradation of anthocyanins.

Figure 3 - Variation of color attributes L* (luminosity), a*, b* (chromatic coordinates), ΔE (overall color difference), h (chromatic tone angle), and C*(chroma) of the FA over 60 days of storage time: light and oxygen presence (LO), light and oxygen absence (A), light presence and oxygen absence (L), and light absence and oxygen presence (O).



Regarding the chromatic behavior of the FB indicator over time, it was observed that the value of the color coordinate L* increased, indicating the lightening of the film after 305 days of storage (Figure 2-b and 4).

Figure 4 - Variation of color attributes L* (luminosity), a*, b* (chromatic coordinates), ΔE (overall color difference), h (chromatic tone angle), and C*(chroma) of the FB over 305 days of storage time: light and oxygen presence (LO), light and oxygen absence (A), light presence and oxygen absence (L), and light absence and oxygen presence (O).



According to the results in Table 1, it can be observed that the color coordinates L*, a*, and b* were influenced by the different storage conditions evaluated, reflected in the ΔE values. Different from the study by Freitas et al. (2020) in which the a* color coordinate was the factor that most influenced the ΔE variation ($p < 0.05$) under different storage conditions evaluated.

Table 1 - Mean values ± standard deviation for the color attributes L*, a*, b*, h, C*, and ΔE of the FB over 305 days in different storage conditions (light and/or oxygen presence or absence).

Color coordinates / Storage Conditions	L*	a*	b*	h°	C*	ΔE
Without storage	10.51±0.98 c	12.79±1.48 b	0.33±0.24 b	1.48±1.09 c	12.80±1.48 b	0.00±0.00 c
305 days in light and oxygen absence (A)	28.87±0.38 b	12.48±1.10 b	0.62±0.35 b	2.78±1.46 c	12.50±1.11 b	18.39±0.37 b

305 days in light and oxygen presence (LO)	32.88±1.55 a	15.85±1.24 a	4.67±1.42 a	16.25±3.89 a	16.55±1.51 a	23.03±1.89 a
305 days in light presence and oxygen absence (L)	31.07±1.29 a	15.90±1.20 a	3.23±1.40 a	11.23±4.14 b	16.25±1.44 a	21.03±1.61 a
305 days in light absence and oxygen presence (O)	29.44±0.49 b	12.13±1.54 b	0.48±0.31 b	2.16±1.25 c	12.14±1.55 b	18.99±0.41 b

L*: Brightness; a*, b*: Chromatic coordinate; ΔE: Global color difference; h°: Chromatic tone angle; C*: Chroma. Means with the same letters in the same column did not differ by the Scott Knott test (α = 0.05). All measurements were carried out in triplicate. Source: Prepared by the authors.

The presence of light was the factor that most affected the color stability of the FB indicator, with the mean values of the attributes L*, a*, b*, C*, and ΔE, under LO and L conditions, being statistically equal ($p > 0.05$), with higher ΔE values. On the other hand, the mean values of the attributes L*, a*, b*, C*, and ΔE of the FB indicators, stored mainly in the absence of light, regardless of the presence of oxygen (A and O), were statistically equal ($p > 0.05$), with smaller colorimetric changes (lower ΔE values) (Table 1), corroborating the result in Figure 2. In the study by Freitas et al. (2020), intelligent cellulose acetate-based films using red cabbage extract also lost color intensity more strongly when they were stored in the presence of light for 30 days.

3.2 Color sensitivity and reversibility of colorimetric indicators

The sensitivity of FB was evaluated amid exposure of the film to basic vapors and of FA amid exposure to acidic vapors. According to a previous study by Gomes et al. (2024), the PVA and gelatin film with EFA presents a complete color change over time under acidic vapors. The PVA and gelatin film with EB exhibited significant differences in color parameters over time under basic vapors (GOMES et al., 2024).

Therefore, the reversibility of FB was evaluated amid exposure to acidic vapors and of FA amid exposure to basic vapors.

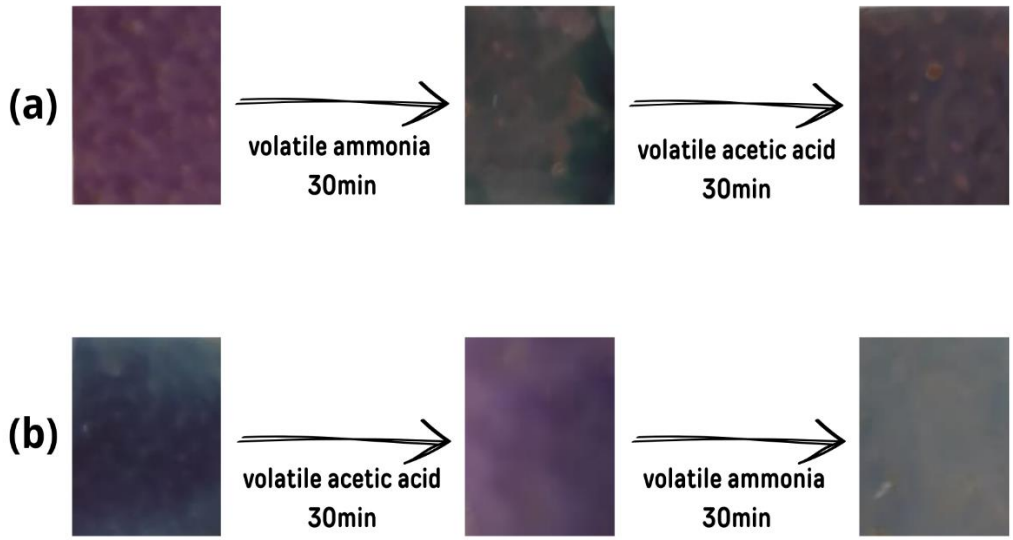
The FB indicator changed color from purple to dark blue/green when exposed to ammonia vapor (30 min), followed by a change from dark blue/green to dark purple after exposure to

acetic acid vapor (30 min) (statistically different values of a*, C*, L*, h° and ΔE, $p < 0.05$, Table 2) and only the color coordinate b* did not differ regarding the chromatic behavior of the indicators ($p > 0.05$). The color change of indicator film prepared with blueberry crude extract for response to ammonia in the Wang et al. (2023) study was very similar.

The FA indicator changed color from blue to purple when exposed to acid vapor (30 min), followed by a change from purple to gray after exposure to ammonia vapor (30 min) (statistically different values of a*, b*, C*, L*, h° and ΔE, $p < 0.05$, Table 2). Indicating the property of sensitivity and partial color reversibility of the materials.

These chromatic transitions and their reversibilities were visually perceptible to the human eye and this result is in agreement with the value determined for the ΔE attribute, which was greater than 3.5 (Table 2) (FERNÁNDEZ-RAMOS et al., 2023; MOKRZYCKI; TATOL, 2011; SCHUBRING, 2002), being that the higher the ΔE value, the more visual the color change of the material (Figure 4) (SCHUBRING, 2002). Thus, FB was more sensitive to basic vapors (ΔE = 15.59) than FA to acid vapors (ΔE = 10.05) and that color reversibility was not fully achieved because the coordinates a*, b*, h° and L* were statistically ($p < 0.05$) different from the initial value for both films (Table 2) and this can be visually verified in Figure 5.

Figure 5 - (a) FB (b) FA color transition under volatile ammonia/acetic acid presence for 30 min and color reversibility under volatile ammonia/acetic acid presence for 30 min.



Shortly after production, the FB indicator showed a coloration with a tendency towards red (a*+) and yellow (b*+), with a significant change ($p < 0.05$) to green (a*-) and blue (b*-), after exposure to volatile ammonia (30 min) (Table 2). Observing the h° the color went from

the first quadrant to the third quadrant, indicating a blue color. This mechanism is related to the interaction between the NH_3 molecules and the water molecules present on the surface and in the polymer chains of the PVA and gelatin film, producing NH_4^+ e OH^-, in which the OH^- increasing the pH of the medium which causes the deprotonation of anthocyanins, causing in the anthocyanin structural changes that promotes the color transition of the films (ZHAI et al., 2017).

Soon after, this same indicator (FB) was exposed to acid vapor (30 min), maintaining the blue color trend (b*-, $p \geq 0.05$), while the color coordinate a* returned to the positive value (a*+, $p < 0.05$), trending towards the red color (Table 2), indicating the partial reversibility of the color attributes and providing a new hue to the indicator (h° went from the third to the fourth quadrant).

Table 2 - Mean values and standard deviation of the color attributes L*, a*, b*, h, C*, and ΔE of the FB under volatile ammonia contact for 30 min and shortly after under volatile acetic acid contact for 30 min (color reversibility), and of the FA under volatile acetic acid contact for 30 min and shortly after under volatile ammonia contact for 30 min (color reversibility).

Films	Condition	L*	a*	b*	h°	C*	ΔE
	Ambient	11.51±0.72 c	12.16±0.52 a	0.13±0.12 a	0.63±0.57 c	12.16±0.52 a	0.00±0.00 c
FB	After in contact with volatile ammonia	20.76±2.25 a	-0.21±0.05 c	-0.92±0.41 b	254.85±7.44 b	0.95±0.39 c	15.59±2.11 a
	After in contact with volatile acetic acid	15.05±1.23 b	7.56±0.14 b	-0.67±0.26 b	354.89±2.07 a	7.59±0.12 b	9.85±2.17 b
FA	Ambient	8.17±1.30 c	-0.27±0.23 c	-3.93±0.12 c	266.02±3.57 c	3.94±0.16 b	0.00±0.00 c
	After in contact with volatile acetic acid	16.03±0.97 b	5.78±1.50 a	-2.68±0.20 a	334.49±4.19 a	6.38±1.43 a	10.05±153 a
	After in contact with volatile ammonia	18.69±1.04 a	3.37±0.53 b	-3.23±0.12 b	316.05±3.50 b	4.67±0.46 b	3.70±2.29 b

Pairs of means followed by at least the same lowercase letter, in the column, do not differ from each other, at 5% significance, using the Scott Knott test. All measurements were carried out in triplicate. Source: Prepared by the authors.

The FA after exposure to acid vapor promoted a significant change ($p < 0.05$) from green (a*-) to red (a*+), also an increase in the b* value remaining negative, tending towards blue coloration, while and h° moved from the third to the fourth quadrant (Table 2). This mechanism is related to the exposure of anthocyanins to the H^+ species released after the interaction of CH_3COOH molecules with water molecules and the polymer chain of the film, which causes the protonation of anthocyanins, favoring the cationic, red form (a*+).

In the reversibility test, the films showed a tendency to approach the original color (Figure 4), but statistically ($p < 0.05$), the color of the films after the reversibility test was different from the color of the original film, without exposure to vapors. The films were not able to return to their original color, possibly due to the various structural changes that occur in the anthocyanins after each stage of exposure to vapors and consequent changes in pH, which may have degraded some of them (ETXABIDE; KILMARTIN; MATÉ, 2021).

Since the films are sensitive to changes in the pH of the medium and are not reversible, it can be concluded that they are capable of permanently recording the quality of food, ensuring that any change is reported and cannot be reversed, in addition to not being susceptible to fraud or adulteration of results. As such, FA and FB films can be tested as colorimetric indicators of fish, which are highly perishable foods and susceptible to enzymatic and microbiological changes, which can produce volatile bases and acids, altering the pH of the food.

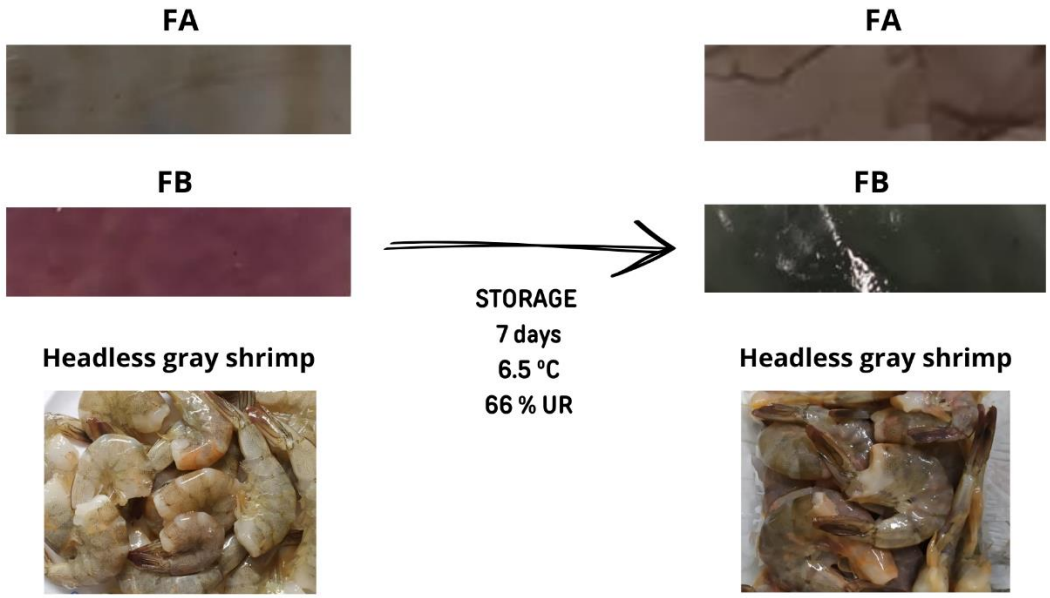
3.3 Evaluation of the colorimetric sensitivity of colorimetric indicators in shrimp

The initial coloration of the fresh raw shrimp was characterized by a grayish and slightly orange shell, indicating the freshness of the food (Figure 6). The degradation of the shrimp was observed visually and through photographic records through the darkening of the crustacean ($\Delta E = 5.99 > 3.5$) (Figure 6), by the odor given off over the storage time, which can be described as ammoniacal, hydrogen sulfide, rancid and indicative of putrefaction, and by the increase in pH from 7.23 ± 0.02 to 7.93 ± 0.02, confirming the formation of ammonia caused by the degradation of proteins and amino acids by enzymatic action and microorganisms, subsequently, with the release of volatile basic nitrogen (JAY; MARIN J. LOESSNER;; DAVID A. GOLDEN., 2005; SALARBASHI et al., 2021), after storage for 7 days under refrigeration (6.5 ºC). According to Normative Instruction No. 23 of August 20, 2019

(MINISTÉRIO DA AGRICULTURA, 2019), the pH of chilled shrimp must be less than 7.85 to be considered quality. Therefore, after 7 days of storage, the food was perceived unfit for consumption.

The values of the colorimetric attributes (L*, a*, b*, C*, h°, and ΔE) of the FA and FB indicators and shrimps, after 7 days of storage under refrigeration (6.5 ºC), can be seen in Table 3. The FA and FB indicators showed colorimetric changes ($p < 0.05$) that impacted the ΔE values of the materials (Table 3), being greater than 3.5, which is considered visible to the human eye, especially for FB that varied from purple to dark green (ΔE = 20.47) and changed the hue angle from the fourth to the second quadrant. However, the FA indicator, in addition to the ΔE of 6.38, changed the hue angle from the second to the first quadrant, reinforcing that the chromatic change of this film can also be visually observed (Figure 6).

Figure 6 - Photographs of the FA and FB indicators and fresh shrimp, before and after storage under refrigeration (6.5 ºC), for 7 days.



This study corroborated the results of Gomes et al. (2024) who also observed that PVA and gelatin-based films, incorporated with crude blueberry extract, were more sensitive than films incorporated with blueberry anthocyanin extract, when exposed to volatile nitrogenous bases, such as those released during shrimp degradation (JAY; MARIN J. LOESSNER;; DAVID A. GOLDEN., 2005).

Previous studies explain the detection mechanism of smart films containing anthocyanins to monitor shrimp freshness, which occurs due to the release of volatile nitrogenous bases, ammonia, trimethylamine, and dimethylamine, from spoiled shrimp. When these volatile

compounds are absorbed by the films, they result in the production of hydroxyl ions that induce the color change of anthocyanins in the polymer matrix (LIU et al., 2021; SALARBASHI et al., 2021; TEIXEIRA et al., 2022a).

On the other hand, since FA indicators change only in the presence of acid vapors, this mechanism does not explain their chromatic behavior. Volatile acids (acetic, propionic, and hydrogen sulfide acids) are also produced by microbial activity during the spoilage of crustaceans and they also cause bad odor in spoiled foods (JAY; MARIN J. LOESSNER;; DAVID A. GOLDEN., 2005). Since PVA and gelatin films added with purified blueberry extract are sensitive to acid vapors, this color change in FA was expected during shrimp degradation (GOMES et al., 2024).

Table 3 - Mean values and standard deviation of the color attributes L*, a*, b*, C*, h, and ΔE of the FB and FA indicator, and shrimps, before and after storage, for 7 days under refrigeration. The comparison was made in pairs (before and after storage).

Conditions / Color coordinates	L*	a*	b*	h°	C*	ΔE
FB Before storage	44.64±5.95 a	9.24±1.48 a	-3.12±2.47 b	342.94±11.52 a	9.88±2.12 a	0.00±0.00 b
FB After storage for 7 days	50.26±6.81 a	-6.51±2.95 b	6.87±1.56 a	131.81±8.73 b	9.54±3.02 a	20.47±2.87 a
FA Before storage	64.70±4.51 a	-2.49±0.91 b	7.73±1.69 a	108.52±9.01 a	8.19±1.49 a	0.00±0.00 b
FA After storage for 7 days	68.98±0.79 a	0.76±0.25 a	8.59±0.97 a	84.78±2.12 b	8.62±0.95 a	6.38±2.59 a
Shirmp Before storage	46.48±4.29 a	3.98±3.40 a	3.69±1.47 a	49.64±16.79 a	5.56±3.38 a	0.00±0.00 a
Shirmp After storage for 7 days	44.66±0.61 a	0.21±0.13 a	2.01±0.65 a	83.65±3.75 a	2.03±0.66 a	5.99±3.53 a

Pairs of means followed by at least the same lowercase letter, in the column, do not differ from each other, at 5% significance, using the t Student test. All measurements were carried out in triplicate. Source: Prepared by the authors.

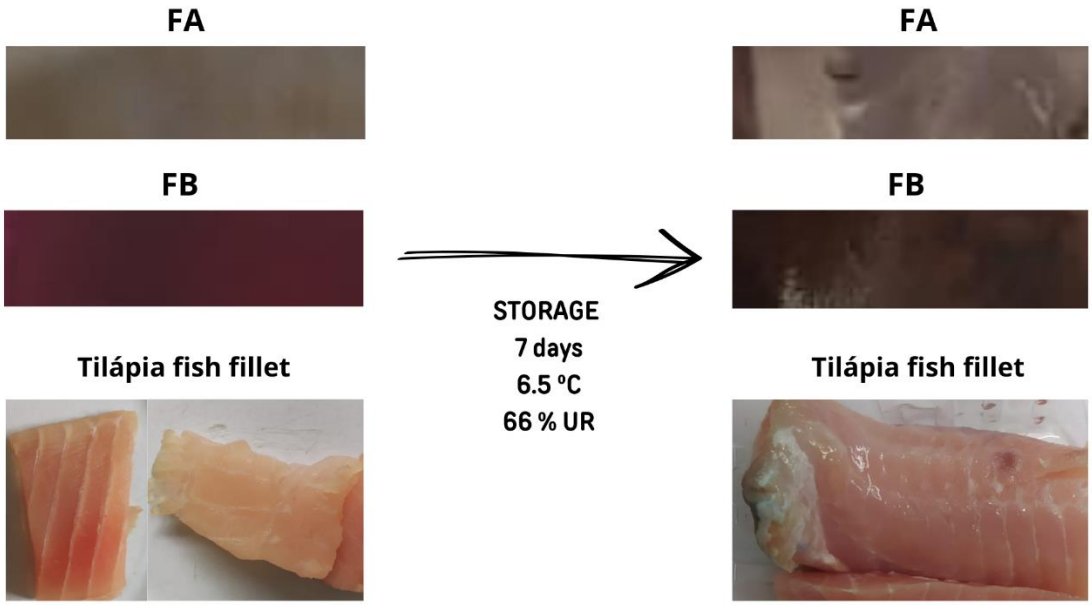
In the study by Teixeira et al. (2022b), the colorimetric indicators of açaí anthocyanin extract, plasticized with glycerol and triethylcitrate, changed from pink to gray, and the ΔE values increased concomitantly with the loss of freshness of the shrimp, indicating the synchrony between the chromatic response and food deterioration, corroborating this study. Other smart packaging has also been developed to monitor shrimp quality, such as pectin films incorporated with curcumin and sulfur nanoparticles (EZATI; RHIM, 2020a) (ΔE = 7.1) and films of chitosan, polyvinyl alcohol, zinc oxide nanoparticles, and anthocyanins from purple potato or roselle, which changed from purple to light green during shrimp storage at 4°C for 8 days (LIU et al., 2021).

Thus, the colorimetric indicators developed from the incorporation of the crude and purified blueberry extract in a PVA and gelatin matrix proved to be an innovative, sustainable, and highly effective solution quickly, accurately, practically, and in real-time monitoring the freshness of refrigerated shrimp, offering an efficient and affordable solution for food quality control, effectively reducing waste.

3.4 Evaluation of the colorimetric sensitivity of colorimetric indicators in Tilapia fillet

The fresh Tilapia fillet changed color from intense reddish to light pink after 7 days of storage under refrigeration (6.5 ºC), as shown in Figure 7, and the values of the colorimetric attributes can be observed in Table 4. The colorimetric difference of the fish over time was visually different since ΔE > 3.5 (ΔE = 6.71), as well as the h° index changed from the first to the second quadrant, indicating greenish tones and loss of quality due to possible oxidation processes or microbial growth during storage.

Figure 7 - Photographs of the FA and FB indicators and fresh tilapia fillet, before and after storage under refrigeration (6.5 ºC), for 7 days.



Fish degradation was also observed by the odor felt during the experiment, which was described as ammoniacal, hydrogen sulfide, rancid, and indicative of putrefaction, also by the increase in pH from 6.18 ± 0.04 to 6.83 ± 0.06. These changes occurred due to the formation of volatile basic nitrogen caused by the degradation of proteins by enzymatic action and microorganisms and the subsequent release of this compound (SALARBASHI et al., 2021). During protein degradation, several volatile nitrogenous bases, such as ammonia, are produced and cause an increase in pH (EZATI; RHIM, 2020c).

According to Decree No. 1255/62 of the Regulation of Industrial and Sanitary Inspection of Products of Animal Origin (RISPOA, 1962), the pH of the external flesh of fresh fish must be less than 6.8 and the pH of the internal flesh must be less than 6.5 to be considered of quality. Thus, before storage, the Tilapia fillet met the legislation criteria (pH = 6.18), although, after storage, the pH values increased to 6.38, being inadaquate according to the legislation, confirming that the fish after 7 days under refrigeration is unfit for consumption.

Once the deterioration of the fresh fish fillet was confirmed, it was observed that both the FB and FA indicators changed their original color ($\Delta E > 3.5$) (Table 4), and can also be used to monitor the quality of the tilapia fillet under refrigeration, indicating that the fish was no longer suitable for consumption after 7 days of storage.

Table 4 - Mean values and standard deviation of the color attributes L*, a*, b*, C*, h, ΔE of the FB and FA indicators and Tilapia fish fillet before and after storage for 7 days. The comparison was made in pairs (before and after storage).

Conditions / Color coordinates	L*	a*	b*	h°	C*	ΔE
FB Before storage	24.57±4.20 a	11.22±4.54 a	-0.92±0.81 b	355.31±4.80 a	11.28±4.52 a	0.00±0.00 b
FB After storage for 7 days	31.22±6.27 a	8.79±1.56 b	4.25±1.18 a	25.92±7.44 b	9.81±1.53 a	9.77±0.78 a
FA Before storage	70.34±3.81 a	-2.27±0.79 b	7.24±0.60 b	107.30±6.04 a	7.62±0.61 a	0.00±0.00 b
FA After storage for 7 days	71.22±3.01a	2.53±1.62 a	8.82±1.06 a	74.78±7.57 b	9.23±1.49 a	6.01±1.04 a
Tilapia fish fillet Before storage	48.94±5.45 a	3.30±3.35 a	6.12±3.56 a	64.36±13.16 b	7.08±4.62 a	0.00±0.00 a
Tilapia fish fillet After storage for 7 days	51.85±2.95 a	-0.96±0.90 a	4.94±0.90 a	101.31±11.05 a	5.09±0.85 a	6.71±4.50 a

Pairs of means followed by at least the same lowercase letter, in the column, do not differ from each other, at 5% significance, using the t Student test. All measurements were carried out in triplicate. Source: Prepared by the authors.

In previous work carried out by the group, it was observed that the PVA and gelatin film, incorporated with crude extract (FB), was more sensitive than the film with purified blueberry extract (FA) to the presence of volatile nitrogenous bases (GOMES et al., 2024). Corroborating what was previously observed for volatile bases, the color difference of FB, before and after storage with tilapia fillet (70 g), was greater (ΔE = 9.77) than that of FA (ΔE = 6.01), allowing visible observation to the human eye of a color change from purple to brown (Figure 6) (h° changed from the fourth quadrant to the first) due to the release of volatile nitrogen during fish degradation over time. The ΔE value of FB was mainly impacted by the color coordinate b* that changed from blue (b* -) to yellow (b*+) (Table 4). On the other hand, the FA indicator

was mainly impacted by the change in hue (h° went from the second quadrant to the first) and by the increase in the a* coordinate, changing the trend from green to red (Table 4), corroborating Figure 7. This color change can be attributed to the sensitivity of the FA indicator to acid vapors that can be produced during the microbiological deterioration of fish and released during storage (GOMES et al., 2024; JAY; MARIN J. LOESSNER;; DAVID A. GOLDEN., 2005).

Some other indicators have already been developed, such as Ezati; Rhim (2020a), who produced a chitosan film with alizarin and was used to monitor the freshness of fish by changing the pH of the fish that exceeded the limit of 7.0 and the color of the indicator film changed from khaki color to light brown color, with a ΔE of 5.8, after 48 h of storage.

The texture of the fish fillet was evaluated by shear force, cohesiveness, and elasticity and presented in Table 5. The cohesiveness is the ratio of the positive force area during the second compression to that during the first compression, representing the internal forces in the food, which keeps the food cohesive (CAINE et al., 2003; GUINÉ et al., 2015). Springiness is a texture parameter that measures the ability of a compressed food to return to its original shape after the applied force is removed; elasticity is an indicator of fresh food (BOURNE, 1982; SAAVEDRA et al., 2022). Storage for 7 days showed no significant differences ($p > 0.05$) in these texture parameters.

In the study by Erfiza (2021), the color of the indicator composed of agar, tapioca, and gambir powder, changed to a darker color during storage of the fish fillet in accordance with changes in pH, but these changes did not agree with changes in the TVB-N value of the fillets. However, the indicators developed in this work have the advantage of not having presented failures after storage of the Tilapia fillet for 7 days (6.5 °C), with the color changes of the films being in accordance with the increase in pH and TVB-N levels and loss of shear force.

The results, thus, demonstrated the innovative nature of the colorimetric indicators, developed by incorporating anthocyanin extract and crude blueberry extract into a PVA and gelatin matrix. This intelligent monitoring technology allows for a practical and real-time visual assessment of the freshness of fish fillets, representing an efficient, sustainable, and affordable solution for food quality control.

4. Conclusion

This study proved that the storage method of colorimetric indicator films is important for the stability of their color and consequently for their proper functionality. The storage time, the

presence of light and oxygen influenced the color stability of the films. Furthermore, it can be concluded that the absence of light provides more stable colorimetric films during storage.

Colorimetric indicators based on PVA and gelatin, produced using raw or purified blueberry extracts, were effective in indicating the freshness of shrimp and tilapia fillets over 7 days of refrigerated storage. These smart indicators are innovative, sustainable, safe for food use, and have great potential to reduce fish waste through simple and rapid communication of food conditions via visual color change.

The color change of FA was less visible to the naked eye when applied to food than *in vitro*, because at the same time that the food produces acid vapors, it also produces basic vapors, and it may also be that there is a smaller release of acidic vapors than basic vapors, hindering the performance of this indicator.

Among the indicators studied, FB stood out for being more stable during storage, sensitive, and irreversible in color change, as well as simpler and faster to develop, with lower process costs since it does not require a purification stage. It also exhibited a more intense colorimetric change and was easier to visualize with the human eye, when tested *in vitro* and applied to monitor fresh tilapia fillets and gray shrimp, food of high-value.

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Declaration of Conflict of Interest

There are no conflicts of interest to declare.

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Figure captions

Figure 1 - Photograph of the system developed for tilapia fillets and shrimp.

Figure 2 - a) FA b) FB photographs shortly after manufactured (T0), after 60 days (FA) or 305 days (FB) in the light and oxygen presence (LO) after 60 days in the light and oxygen absence (A), after 60 days (FA) or 305 days (FB) in the light presence and oxygen absence (L) and after 60 days (FA) or 305 days (FB) in the light absence and oxygen presence (O).

Figure 3 - Variation of color attributes L* (luminosity), a*, b* (chromatic coordinates), ΔE (overall color difference), h° (chromatic tone angle), and C*(chroma) of the FA over 60 days of storage time: light and oxygen presence (LO), light and oxygen absence (A), light presence and oxygen absence (L), and light absence and oxygen presence (0).

Figure 4 - Variation of color attributes L* (luminosity), a*, b* (chromatic coordinates), ΔE (overall color difference), h° (chromatic tone angle), and C*(chroma) of the FB over 305 days of storage time: light and oxygen presence (LO), light and oxygen absence (A), light presence and oxygen absence (L), and light absence and oxygen presence (0).

Figure 5 - (a) FB (b) FA color transition under volatile ammonia/acetic acid presence for 30 min and color reversibility under volatile ammonia/acetic acid presence for 30 min.

Figure 6 - Photographs of the FA and FB indicators and fresh shrimp, before and after storage under refrigeration (6.5 ºC), for 7 days.

Figure 7 - Photographs of the FA and FB indicators and fresh tilapia fillet, before and after storage under refrigeration (6.5 ºC), for 7 days.

6. GENERAL CONCLUSION

Gelatin and PVA polymer blends with blueberry extracts show potential as smart packaging material due to their thermal stability, good thickness, flexibility, moderate resistance and reduced sensitivity to water.

Among the films developed, the one with blueberry phenolic extract did not demonstrate the greatest potential as a colorimetric indicator. The PVA and gelatin film with crude blueberry extract (FB) was the most sensitive to ammonia vapor, indicating its suitability for monitoring the shelf life of foods that release basic vapors during spoilage. Meanwhile, the film incorporated with anthocyanin extract (FA) was the most sensitive to acetic acid vapor, being recommended for colorimetric indication when exposed to volatile acids.

Both FB and FA effectively identified spoilage after seven days of storage of fresh tilapia fillets and gray shrimp, which release acids and volatile nitrogenous basic compounds during degradation, being an excellent sustainable alternative to monitor the quality and freshness of these high-value foods and reduce waste. However, FB stands out as the superior intelligent indicator due to its greater stability during storage, high sensitivity, and irreversible color change. Its is also simpler and more cost-effective to produce, as the crude extract does not require purification. FB presented a more intense colorimetric change that was clearly perceptible to the naked eye, making it an easy-to-use tool for consumers to ensure food safety.

This study brought a scientific contribution to the technical approach, and brought an innovation because the color indicator offers an easier, more visual way to ensure consumer safety. With the colorimetric indicator, it is possible to know the quality of the food without needing to see the product that is packaged, as the packaging is often opaque and does not allow the food to be seen on the market. The FB color indicator has proven to be socially applicable, with the end consumer being its main beneficiary.

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APPENDIX A

Supplementary material for chapter 2 of article 1: Gelatin/polyvinyl alcohol films incorporated with different blueberry extracts as potential colorimetric indicators to detect acidic and basic vapors

Tabela 1 - p-value of the color attributes (L*, a*, b*, ΔE, h° and C*) of films incorporated with crude, phenolic and anthocyanin extracts after exposure for 28 min to acid vapors.

Variation factor	**p-value**					
	L*	a*	b*	ΔE	h°	C*
Extract type (E)	0.00*	0.00*	0.00*	0.00*	0.00*	0.00*
Time (T)	0.67 ns	0.00*	0.31 ns	0.00*	0.00*	0.88 ns
E x T	0.28 ns	0.00*	0.37 ns	0.00*	0.00*	0.16 ns

F test at 5% significance level. p: probability of 5%. *significant (p≤0.05). ns: not significant (p>0.05). L*: Brightness; a*, b*: Chromatic coordinate; C*: Chroma; h°: Chromatic tone angle; ΔE: Global color difference. Source: Prepared by the authors.

Tabela 2 - Mean values and standard deviation of the color attributes L*, b* and C* of the films depending on the type of extract added, after exposure to acid vapors.

	Extract type	**Mean values**
L*	Phenolics	82.78 ± 1.11 a
	Crude	69.73 ± 0.97 b
	Anthocyanin	53.19 ± 2.71 c
b*	Crude	21.96 ± 0.99 a
	Phenolics	13.49 ± 2.43 b
	Anthocyanin	9.39 ± 0.98 c
C*	Crude	23.21 ± 1.11 a
	Phenolics	13.67 ± 2.43 b
	Anthocyanin	11.08 ± 1.33 c

Data expressed as mean ± standard deviation. Pairs of means followed by at least one vertical lowercase letter do not differ from each other, at 5% significance, using the Tukey test. L*: Brightness; b*: Chromatic coordinate; C*: Chroma. Source: Prepared by the authors.

Tabela 3 - Mean values and standard deviation of the color attributes a*, hº and ΔE of the films depending on the type of extract added and the time of exposure to acid vapors.

	Extract type	**Phenolics**	Crude	Anthocyanin
	Time			
h°	0	81.64 ± 1.41 b A	72.20 ±1.39 b A	101.53 ±10.59 a A
	4	80.81 ± 2.24 a A	71.51 ± 2.05 ab A	70.32 ± 6.09 b B
	8	81.23 ± 2.34 a A	72.42 ± 3.35 ab A	70.03 ± 11.84 b B
	12	80.79 ± 2.17 a A	72.06 ± 1.40 ab A	65.25 ± 6.17 b BC
	16	80.28 ± 1.61 a A	69.08 ± 3.43 b A	57.48 ±10.32 c BC
	20	82.53 ± 1.56 a A	71.07 ± 0.16 b A	43.41 ± 7.16 c D
	24	79.95 ± 2.09 a A	71.21 ± a 0.82 aA	53.82 ± 3.14 b CD
	28	80.46 ± 2.01 a A	70.68 ± a 0.16 A	53.11 ± 9.00 b CD
ΔE	0	0.00± 0.00 a C	0.00 ± 0.00 a A	0.00 ± 0.00 a D
	4	3.11 ± 1.05 b BC	3.03± 0.61 b A	9.39 ± 2.41a BC
	8	3.47±2.63 ab BC	1.93 ± 0.64 b A	6.22 ± 1.77 a C
	12	5.47 ± 1.65 b AB	2.37 ± 1.40 b A	9.49 ± 2.92 a BC
	16	6.77 ± 1.45 b AB	4.37 ± 1.36 b A	10.55 ± 2.71a ABC

	20	8.43 ± 2.48 b A	2.57 ± 1.66 c A	14.36 ± 1.38 a A
	24	6.36 ± 1.69 b AB	2.19 ± 1.27 c A	11.78 ± 2.97 a AB
	28	5.69 ± 2.84 b AB	2.45 ± 1.06 c A	10.31 ± 0.84 a ABC
a*	0	2.62 ± 0.12 b A	6.78 ± 1.69 a A	-1.96 ± 1.44 c C
	4	2.38 ± 0.41 b A	7.82 ± 1.83 a A	3.29 ± 1.48 b B
	8	2.31 ± 0.76 c A	6.81 ± 2.03 a A	3.43 ± 2.44 b B
	12	2.03 ± 0.36 b A	7.49 ± 1.77 a A	5.38 ± 2.49 a B
	16	1.97 ± 0.08 b A	8.66 ± 0.63 a A	6.19 ± 2.00 a AB
	20	1.35 ± 0.30 b A	7.29 ± 0.53 a A	9.43 ± 2.26 a A
	24	2.18 ± 0.42 b A	7.47 ± 1.01 a A	6.59 ± 2.75 a AB
	28	2.09 ± 0.43 b A	7.42 ± 0.46 a A	6.65 ± 1.90 a AB

Data expressed as mean ± standard deviation. Pairs of means followed by at least the same lowercase letter horizontally and at least one capital letter vertically do not differ from each other, at 5% significance, using the Tukey test. a*: Chromatic coordinate; h°: Chromatic tone angle; ΔE: Global color difference. Source: Prepared by the authors.

Tabela 4 - p-value of the color attributes (L*, a*, b*, ΔE, h° and C*) of films incorporated with crude, phenolic and anthocyanin extracts after exposure for 28 min to basic vapors.

Variation factor	**p-value**					
	L*	a*	b*	ΔE	h°	C*

Extract type (E)	0.00*	0.00*	0.00*	0.00*	0.00*	0.00*
Time (T)	0.00*	0.00*	0.00*	0.00*	0.00*	0.00*
E x T	0.17 ns	0.00*	0.00*	0.00*	0.00*	0.00*

F test at 5% significance level. p: probability of 5%. *significant ($p \leq 0.05$). ns: not significant ($p > 0.05$). L*: Brightness; a*, b*: Chromatic coordinate; C*: Chroma; h°: Chromatic tone angle; ΔE: Global color difference. Source: Prepared by the authors.

Tabela 5 - Mean values and standard deviation of the L* coordinate of the films depending on the type of extract added and the time of exposure to ammonia.

	Extract type	**Média**
L*	Phenolics	79.95 ± 1.15 a
	Crude	71.56 ± 2.25 b
	Anthocyanin	53.30 ± 0.92 c
	Time (min)	
	0	70.49 ± 13.87 a
	4	68.94 ± 14.43 ab
	8	68.36 ± 14.73 abc
	12	68.28 ± 13.75 abc
	16	68.36 ± 13.62 abc
	20	68.19 ± 12.97 abc
	24	67.17 ± 13.39 bc
	28	66.36 ± 12.45 c

Data expressed as mean ± standard deviation. Pairs of means followed by at least one vertical lowercase letter do not differ from each other, at 5% significance, using the Tukey test. L*: Brightness. Source: Prepared by the authors.

Tabela 6 - Mean values and standard deviation of the color attributes a*, b*, C*, hº and ΔE of the films after exposure to volatile bases.

	Extract type / **Time**	**Phenolics**	Crude	Anthocyanin
h°	0	82.71 ± 0.53 b A	69.52 ± 2.86 c C	153.05 ± 5.68 a B
	4	88.63 ± 1.58 b A	72.81 ± 2.27 c BC	167.75 ± 9.59 a A
	8	87.81 ± 3.39 b A	78.86 ± 4.03 c ABC	171.52 ± 3.79 a A
	12	85.46 ± 2.80 b A	81.59 ± 4.15 b AB	172.23 ± 2.27 a A
	16	87.38 ± 3.56 b A	84.57 ± 4.40 b A	165.97 ± 3.67 a A
	20	86.89 ± 0.64 b A	83.92 ± 3.74 b A	168.05 ± 9.72 a A
	24	86.58 ± 3.40 b A	85.57 ± 1.29 b A	161.97 ± 7.37 a AB
	28	88.77 ± 1.51 b A	88.45 ± b 0.75 A	162.06 ± 4.38 a AB
a*	0	1.95 ± 0.22 b A	6.48 ± 0.40 a A	-6.82 ± 0.43 c A
	4	0.62 ± 0.71 b A	5.45 ± 1.36 a AB	-12.51 ± 0.89 c B
	8	0.97 ± 1.48 b A	4.08± 1.40 a ABC	-13.53 ± 0.86 c B
	12	2.26 ± 1.11 a A	3.37 ±1.70 a BCD	-13.35 ± 0.44 b B

	16	1.18 ± 1.55 a A	2.27 ± 1.79 a CD	-12.42 ± 0.54 b B
	20	1.58 ± 0.25 a A	2.73 ±1.74 a BCD	-12.99 ± 0.64 b B
	24	1.73 ± 1.70 a A	2.13 ± 0.61 a CD	-12.81 ± 0.97 b B
	28	0.55 ± 0.66 a A	0.72 ± 0.30 a D	-12.38 ± 0.67 b B
ΔE	0	0.00 ± 0.00 a B	0.00 ± 0.00 a D	0.00 ± 0.00 a B
	4	10.12 ± 3.20 a A	4.29 ± 2.96 b CD	6.25 ± 0.92 ab A
	8	11.55 ± 0.79 a A	5.88 ± 1.76 b C	7.69 ± 0.32 ab A
	12	14.62 ± 3.03 a A	7.59 ± 3.14 b BC	6. 97 ± 0.21 b A
	16	12.44 ± 1.26 a A	8.81 ± 0.37ab ABC	6.67 ± 1.54 b A
	20	14.27 ± 2.48 a A	9.24 ± 3.86 b ABC	6.60 ± 1.51 b A
	24	13.67 ± 3.94 a A	12.23 ± 2.38 a AB	6.72 ± 0.96 b A
	28	11.31 ± 0.55 a A	13.42 ± 0.79 a A	5.81 ± 0.29 b A
b*	0	15.36 ± 2.53 a B	17.47 ± 1.62 a C	3.46 ± 0.65 b A
	4	25.29 ± 0.86 a A	17.49 ± 2.64 b C	2.68 ± 1.99 c A
	8	26.67 ± 1.77 a A	20.91 ± 3.14 b BC	2.02 ± 0.94 c A
	12	29.85 ± 3.50 a A	22.67 ± 0.41 b ABC	1.82 ± 0.55 c A
	16	27.63 ± 2.27 a A	24.04 ± 1.30 a AB	3.14 ± 0.96 b A

	20	29.31 ± 2.34 a A	24.87 ± 2.08 b AB	2.72 ± 2.20 c A
	24	28.65 ± 1.65 a A	27.56 ± 0.47 a A	4.13 ± 1.56 b A
	28	26.00 ± 3.17 a A	27.06 ± 1.98 a A	3.99 ± 0.86 b A
C*	0	15.49 ± 2.53 a C	18.65 ± 1.39 a D	7.67 ± 0.17 b B
	4	25.31 ± 0.87 a B	18.33 ± 2.87 b D	12.91 ± 0.45 c A
	8	26.71 ±1.71 a B	21.34 ± 3.09 b CD	13.70 ± 0.90 c A
	12	29.96 ± 3.39 a A	22.96 ± 0.54 b BCD	13.48 ± 0.45 c A
	16	27.69 ± 2.18 a AB	24.19 ± 1.23 a ABC	12.83 ± 0.75 b A
	20	29.35 ± 2.33 a AB	25.05 ± 2.20 b ABC	13.40 ± 0.27 c A
	24	28.73 ± 1.66 a AB	27.65 ± 0.46 a A	13.53 ± 0.54 b A
	28	26.01 ± 3.17 a AB	27.071 ± 1.97 a AB	13.03 ± 0.42 b A

Data expressed as mean ± standard deviation. Pairs of means followed by at least the same lowercase letter horizontally and at least one capital letter vertically do not differ from each other, at 5% significance, using the Tukey test. a*, b*: Chromatic coordinate; C*: Chroma; h°: Chromatic tone angle; ΔE: Global color difference. Source: Prepared by the authors.

APPENDIX B

Supplementary material for chapter 4 of article 3: Stability and reversibility of colorimetric indicators, and their sensitivity in the presence of packaged tilapia and shrimp fillets

B 1. Method the monitoring the degradation of Tilapia fillets

The monitoring of the degradation of fresh Tilapia fillets during the storage was performed by determining the pH (2.2.8), the total volatile base index (TVB-N), the sensory characteristics of odor, color (2.2.5) and texture. The analyses were performed in 3 replicates, in two times: T0, on the day of collection of the food at the local market and T7, after 7 days of storage.

The total volatile nitrogen base (TVB-N) was determined according to Adolfo Lutz Institute (2005) in tilapia fillet immediately after acquisition from the market and after 7 days of storage under refrigeration (6.5 ºC ± 2.0 ºC / RH 66 ± 2.0 %), in duplicate. 10 g of sample, 2 g of magnesium oxide, 300 mL of water, and some pieces of ceramic, used as an antifoaming agent, were added to the distillation flask and coupled to a distiller (model M25008, Quimis, Brazil). The distillate was collected in a bottle containing 15 mL of 0.05 M aqueous sulfuric acid solution and 0.04 mL of 0.02% methyl red indicator. Then, the obtained solution was titrated with 0.1 M sodium hydroxide until the color changed from red to yellow. The amount of TVB-N, expressed in mg.100 g-1 of tilapia sample, was calculated using the volume (V) of sulfuric acid added and its concentration (C) according to Equation 4:

$$TVBN(mg.100g^{-1}) = \frac{(V1-V2)x\ f1x\ 0{,}0014}{p\ x\ f2}\ x\ 100 \quad (4)$$

in which V1 is the volume of sulfuric acid (mL), V2 is the volume of NaOH (mL), N is the normality of HCl (0.1 N), f1 and f2 are the correction factors of sulfuric acid and sodium hydroxide solution, respectively and p is the weight of the fish sample (g).

Texture profile analysis was measured according to the method of Peng et al. (2023) and Saavedra et al. (2022), with modifications, in fish after collection from the market and after 7 days of storage. The dorsal muscle of tilapia fillet was cut into cubes of 20 mm × 20 mm × 20 mm. The Universal Mechanical Testing Machine (Instron Corporation, Norwood, MA, USA) equipped with a blade (diameter 15 mm) was used to press the cubes perpendicular to the direction of the muscle fibers at a speed of 60 mm/min. The following parameters were

determined: elasticity (mm), cohesiveness (dimensionless), shear force (N). Each determination was performed 10 times.

B 2. Result the monitoring the degradation of Tilapia fillets

Fish degradation was also observed by the odor felt during the experiment, which was described as ammoniacal, hydrogen sulfide, rancid and indicative of putrefaction, by the increase in pH from 6.18 ± 0.04 to 6.83 ± 0.06 and TVB-N level from 44 mg N/100 g to 74 mg N/100 g.

Despite the increase in the content of volatile nitrogen compounds and the evidence that the fish exceeded the TVB-N limit permitted by law after 7 days of storage, the fresh fish fillet before storage did not meet the maximum limit permitted of 30 mg of nitrogen/100 g by Ordinance No. 185 of 1997 (Ministério da Agricultura e Pecuária, 1997). This result may have occurred due to the perishability of the fish fillet which, despite having been purchased at the local market on the day of receipt, it may have suffered some degradation during previous transportation and storage.

Shear force is the force applied to food that causes it to deform or break. The firmer the food, the greater the shear force (PENG et al., 2023). In this study, the shear force decreased significantly ($p<0.05$) after the degradation of the fish, indicating loss of its firmness and presentation of a sticky texture indicating deterioration. The loss of firmness of fish may be mainly related to changes in purine metabolism, amino acid metabolism, and choline metabolism pathway caused by oxidative stress, destroying the cellular structure and ultrastructure of the fish's muscle fiber (PENG et al., 2023). Similar results were found in the work of Shi et al. (2020), in which the shear force of catfish fillets under 1 h, 8 h and 24 h of pre-cooling decreased continuously with the extension of storage duration.

Table 5 - Mean values and standard deviation of the texture parameters (Shear force, Cohesiveness and Springiness) of the Tilapia fish fillet before and after storage for 7 days.

Conditions the Tilapia Fish fillet / Texture parameters	Shear force (N)	Cohesiveness (dimensionless)	Springiness (mm)
Before storage	9.40±2.24 a	0.66±0.18 a	7.50±1.51 a

After storage for 7 days	4.91±0.65 b	0.68±0.10 a	6.29±1.15 a

Pairs of means followed by at least the same lowercase letter, in the column, do not differ from each other, at 5% significance, using the t Student test. All measurements were carried out in triplicate. Source: Prepared by the authors.

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